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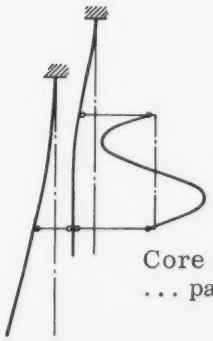
REACTOR AND FUEL-PROCESSING TECHNOLOGY

A Quarterly Technical Progress Review

Prepared for DIVISION OF TECHNICAL INFORMATION, U. S. ATOMIC ENERGY COMMISSION,
by ARGONNE NATIONAL LABORATORY



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● VOLUME 10

● NUMBER 3

TECHNICAL PROGRESS REVIEWS

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Reactor and Fuel-Processing Technology

With this third issue of the merged Technical Progress Reviews *Power Reactor Technology* and *Reactor Fuel Processing*, the name is being shortened to *Reactor and Fuel-Processing Technology*.

Reactor and Fuel-Processing Technology is a quarterly review of developments in reactor and reactor fuel technology. As such, this journal reports and interprets progress in the reactor field in terms of its significance to the reactor designer, operator, and fuel-cycle specialist. Articles either summarize and critically evaluate recently reported developments or review the "state of the art" of a particular topic. Both types of articles reference reports and publications that merit study; readers are urged to consult the references for additional information and the judgments of the original authors. If a reader has information that causes his evaluation to reinforce, modify, or contradict the opinions of our reviewers, he is encouraged to write to the Editor.

The scope of the journal encompasses:

CONCEPTS and APPLICATIONS: Progress in evaluating the applicability and economics of various reactor types and systems (including unconventional approaches), as well as of fuel resources and cycles, for utility central-station generation of electricity, auxiliary power, process radiation and heat, desalting, and propulsion—and for terrestrial, undersea, aerospace, and other advanced uses.

ANALYSIS and EXPERIMENTATION: Advancements in the techniques of reactor physics, fluid and thermal technology, energy conversion, fuel elements, materials, mechanics, control and dynamics, and reactor safety.

SYSTEMS and COMPONENTS: Experience as reflected in design and construction practice, components, systems technology, and operating performance of various specific types of reactors—including pressurized- and boiling-water reactors, molten-salt, organic-, gas-, and liquid-metal-cooled reactors, as well as generally applicable aspects of research and test reactors.

FUEL PROCESSING: Recent research and development on fuel aqueous processing, nonaqueous processing, waste disposal, and processing safety.

This journal is prepared by Argonne National Laboratory at the request of the Division of Technical Information of the U. S. Atomic Energy Commission. At Argonne, the Reactor Engineering Division (L. J. Koch, Director) and the Chemical Engineering Division (R. C. Vogel, Director) have the principal responsibility for the preparation of *Reactor and Fuel-Processing Technology*, with the regular assistance of the Reactor Physics Division (R. A. Avery, Director) and the Idaho Division (M. Novick, Director) and the occasional assistance of other divisions (Metallurgy, Chemistry, and Reactor Operations). Thus, unless noted otherwise, the reviewers and authors of articles are Laboratory staff members. The editors welcome interpretive review articles contributed by authors outside the Laboratory.

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Argonne National Laboratory

Small Utility Nuclear Power Plants— Their Performance and Market Potential

By John F. Marchaterre

A three-part study reexamining the status and potential of small nuclear power plants [<100 Mw(e)] for utility use has been completed by AEC.[1-3] Because the study will be of considerable interest to reactor designers and operators, as well as to the managements of small utilities, this article summarizes the results of the study.

The first part of the study presents a compilation of design, construction, and operating information for 10 existing small nuclear plants through 1965. The second part, which is based on information from reactor suppliers, describes the technical and economic performance of small nuclear plants that are being offered now or being planned for the future. The third part of the study is an assessment by Kaiser Engineers of the general and economic status of small nuclear plants; it also examines areas of potential cost reductions and evaluates courses of action necessary to achieve competitive status and acceptance.

Experience with 10 Plants

Of the 10 plants surveyed, 4 have boiling-water reactors (Big Rock Point Nuclear Power Plant, Elk River Reactor, Humboldt Bay Power Plant, and La Crosse Boiling Water Reactor), two have boiling-water reactors with integral nuclear superheaters [Boiling Nuclear Superheat (BONUS) Reactor and Pathfinder Atomic Power Plant], and there is one heavy-water-cooled and -moderated pressure-tube reactor (Carolinas-Virginia Tube Reactor), an organic-cooled and -moderated reactor (Piqua Nuclear Power Facility), a sodium-cooled graphite-moderated reactor (Hallam Nuclear Power Facility), and a gas-cooled reactor (Peach Bottom Atomic Power Station).

Tables 1 to 3 summarize the data on these 10 plants. As of December 1965, 6 of the 10 plants had operated; BONUS, Pathfinder, Peach Bottom, and La Crosse had not. Although more recent operating experience has been reported,[4] and Big Rock Point and Humboldt Bay experience has been described in connection with larger plants,[5] the as-of-December-1965 compilation in the first part of the study is particularly worth considering in context because of the perspective it gives to the projections in the second and third parts of the study. The operational experiences of the six plants that had operated by December 1965 are summarized below.

Big Rock Point. The 157-Mw(t) rated power of this advanced boiling-water reactor was achieved on Mar. 21, 1963. Normal operation was intended to be delayed by initial operations as part of a planned research and development program. However, normal operation was further delayed by two reactor problems that required about 15 months of downtime to accomplish major reactor modifications.[5-7] The plant began normal operation in September 1965 and was performing reliably in December 1965. As of Apr. 30, 1966, electricity generation totaled 632,864,900 kw-hr (net).

Elk River. This boiling-water-reactor plant achieved full power on Feb. 10, 1964. Operation during the first year was interrupted numerous times by operating problems and control-rod inspections.[4,7,8] The plant began reliable operation in March 1965; through April 1966, it had generated 253,206,036 kw-hr (net) of electricity.

Humboldt Bay. This boiling-water-reactor plant achieved full power on May 12, 1963. After an initial 2.5-month period when the plant was shut down for modifications, the plant produced 818,340,000 kw-hr of electricity through December 1965 and achieved availability and capacity factors of 0.88 and 0.83, respectively.

Carolinas-Virginia Tube Reactor. This heavy-water-cooled and -moderated reactor plant achieved full power on May 7, 1964, and commercial operation was started simultaneously. With only minor difficulties, the plant had generated 166,237,000 kw-hr

Table 1 FACILITY DATA[1]

	La Crosse	Big Rock	Humboldt Bay	Elk River*	Pathfinder*	BONUS*	CVTR*	Hallam	Peach Bottom	Piqua
Acronym	LACBWR			ERR	CRBR	BONUS	CVTR	HNPF	HTGR	PNPF
Location	Genoa, Wis.	Charlevoix, Mich.	Eureka, Calif.	Elk River Minn.	Sioux Falls, S. Dak.	Puerto Rico	Parr, S. C.	Hallam, Nebr.	Peach Bottom, Pa.	Piqua, Ohio
Owner	AEC	Consumers Power Co.	Pacific Gas & Electric Co.	AEC	Northern States Power Co.	AEC	Carolinas-Va. Nuclear Power Assoc.	AEC	High Temp. Reactor Development Assoc.	AEC
Operator	Dairyland Power Coop.	Same	Same	Rural Coop. Power Assoc.	Same	Puerto Rico Water Resources Authority	Same	Consumers Public Power District†	Philadelphia Electric Co.	City of Piqua
Reactor vendor	A-C†	GE	GE	A-C†	A-C†	GNEC†	West.	AI†	GA	AI†
Architect-engineer	Sargent & Lundy	Bechtel†	Bechtel†	Sargent & Lundy	Pioneer Service & Eng.	Jackson & Moreland	Stone & Webster	Bechtel†	Bechtel†	Holmes & Narver
Reactor type	BWR	BWR	BWR	BWR	BWR integral superheat	BWR integral superheat	Pressure tube D_2O	SGR	Gas cooled	Organic moderated & cooled
Status, 1/1/66	In construction	Operating	Operating	Operating	Operating	Operating	Operating	Shut down	In startup	Operating
Initial power, Mw(e) net	52.4	48.5	50.2	22.5	62.0	16.5	11.3	75.0	40	11.4
Mw(t)	165	157	165	58.2/14.8	157.2/42.4	37/13	47.2/13	240	115	45.5
Stretch power, Mw(e) net		72.8	68				17.0			
Mw(t)		240	240	116			65/17			
Stretch achieved?	N.A.‡	Yes	Yes	No	N.A.	N.A.	Yes	N.A.	N.A.	N.A.
Moderator	H_2O	H_2O	H_2O	H_2O	H_2O	H_2O	D_2O	Graphite	Graphite	Terphenyl
Coolant	H_2O	H_2O	H_2O	H_2O	H_2O/steam	H_2O/steam	D_2O	Sodium	He	Terphenyl
Power generated as of 8/31/65, 10^6 kw-hr (gross)	0	334.0	798.0	144.4	0	2.519	86.95	192.46	0	55.91
Comment					First superheater core at 31.5 Mw(t)		61.8 Mw(t) from fuel, 3.2 Mw(t) from moderator	Operation up to 256 Mw(t) has been achieved		

*Boiler/superheater. †Prime contractor(s). ‡Not applicable.

Table 2 REACTOR CORE DATA[1]

	La Crosse	Big Rock	Humboldt Bay	Elk River	Pathfinder*	BONUS*	CVTR	Hallam	Peach Bottom	Piqua
Thermal power,† Mw(t)	165	240	240	58.2	199.6	50	65	240	115	45.5
Fuel Material	UO_2	UO_2	UO_2	UO_2-ThO_2	UO_2/UO_2 cermet	UO_2	UO_2	U–10 Mo	UO_2-ThC_2-C	U–3.5 Mo
Fuel shape	Rods	Rods	Rods	Rods	Rods/tubes	Rods	Rods	Rods	Rods	Concentric finned tubes
Fuel cladding	348H SS‡	304 SS	304 SS	304 SS	Zr-2/316L SS	Zr-2/Inconel	Zr-4	304 SS	Graphite	Al
No. of bundles currently used	72	84	172	148	96/415	64/32	72	140	804	70
Fuel loading, ceramic, metric tons	9.75	12.7	18.9	4.35 ThO_2 0.21 UO_2	7.45/0.051	3.18/2.03	3.18	27.6§	1.60 ThC_2 0.26 UC_2	5.76§
Enrichment, %	3.63	2.6–4.2	2.57	93.13	2.2–3.2/93	2.4/3.25	1.5–2.0	3.6	93	1.94
Core 1 design burnup, Mwd/metric ton	14,000	16,500	11,000	9500	11,000/185,000	11,000/6850	13,400		60,000	3000
Core 1 life, full-power days	738	777	920	655	465/196	845/954	586		877	380
Power density, av. kw/liter	41.4	46	39.7	21.0	45/46	33.6/11.5	7.6	4.8	8.3	14.4
Specific power, av. kw/kg	17.0	18.9	12.7	12.7	21.1/830	11.6/6.4	20.8	7.8¶	61.8	7.6¶
Heat flux, av. 10^3 Btu/(hr)(sq ft)	109.1	116	118	90.7	128/77.8	106/66	129	165	71.0	42
No. of control rods	29	32	32	13	16/48	9/8	28	19	36	13
Poison material	B_4C	B_4C & SS	B_4C	Boron SS	Boron SS	Boron SS	SS & boron SS	Gd–Sm	B_4C	B_4C
Rod drive	Roller–screw	Hydraulic piston	Hydraulic piston	Rack–pinion	Rack–pinion	Rack–pinion	Rack–pinion	Ball–screw	Ball–screw	Magnetic jack
Drive location	Below	Below	Below	Below	Above	Above	Above	Above	Below	Above
Vessel size, height (ID), in.	444(87)	360(106)	504(120)	300(84)	433(132)	319(84)	132(111)	396(228)	426(168)	324(91)
Core size, length (dia.), in.	83(61)	70(76.5)	79(84)	60(60)	72(69)	55(58)	96(83)	159(156)	90(109)	60(64)
Comment		Core 2: Zr-2	Core 2: Zr-2 and 15,400 Mwd/metric ton of U	Rod material may be replaced with B_4C; core 2: 348 SS			12 boron SS and 16 SS control rods used	UC fuel for core 2	Uses 19 emergency & 55 shutdown rods	Core 2 will be SAP-clad UC

*Boiler/superheater. †Power for which data are presented. ‡SS = stainless steel; Zr-2 = Zircaloy-2. §Uranium only. ¶kw/kg U + Mo.

Table 3 PLANT COSTS IN DOLLARS (EXCEPT ENERGY COST IN MILLS PER KILOWATT-HOUR)[1]

	La Crosse	Big Rock Point	Humboldt Bay	Elk River	Pathfinder	BONUS	CVTR	Hallam	Peach Bottom	Piqua
Land and land rights	102,000	183,000			366,304	148,000	67,000		341,000	4,000
Direct construction cost										
Structures and improvements	3,087,000*	3,324,333	4,175,000	1,765,837	3,370,000	3,146,814	4,100,000	13,034,015	2,511,385	2,239,000
Nuclear, less core	10,404,000	9,214,082	10,375,000	3,254,739	8,348,000	4,257,094	13,154,000	14,543,454	14,052,185	5,213,000
Electrical equipment	304,000	650,951	705,000	282,101	364,000	498,249	1,443,000	14,863,066	870,500	195,000
Turbogenerator	2,974,000	4,007,534	4,095,000	1,121,842	6,740,000	1,405,450			4,400,000	
Transmission plant	165,000	450,035		134,114		145,600			422,500	
Other	2,051,000	523,878	460,000	169,136	325,000	305,611	579,000		1,127,115	500,000
Subtotal (direct cost)	18,985,000	18,170,813	19,810,000	6,727,769	19,147,000	9,758,818	19,276,000	42,420,535	23,383,685	8,151,000
Indirect construction cost†										
Interest during construction		881,538		2,432,819	3,486,935			6,290,588	651,000	
Engineering and design				1,632,452		5,486,617		6,045,888	2,961,315	
General and administrative		6,961,598		468,973	2,663,674	2,194,530		1,198,501	487,000	
Other				2,476,487	134,209	572,389		4,385,134	275,000	
Subtotal (indirect cost)		7,843,136	4,390,000	7,010,728	6,284,818	8,253,536		17,920,111	4,374,315	
Contingencies				701,000		12,146				
Total construction cost	19,087,000	26,196,949	24,200,000	14,439,497	25,798,122	18,024,500	19,343,000	60,340,646	28,099,000	8,151,000
Core 1 fuel cost	638,000	1,695,084	~2,000,000	417,000	3,160,000	1,515,236	1,405,000	2,732,084	1,000,000	1,243,159
Energy cost, mills/kw-hr										
Fuel	3.10	2.7	4.07	3.5–4.21	4.32			4.85		3.86–4.71
Operating and maintenance	1.72	1.8	1.44	2.87–3.42	1.23			1.34		1.49–8.80
Fixed charges	3.63	7.5	6.30	7.06–7.2	8.61			14.71		3.41–5.24
Total	8.45	12.0	11.78	13.43–14.83	14.16	25–30		20.90		8.76–18.75

*Nonreactor plant structures and improvements only.
†La Crosse, CVTR, and Piqua indirect construction costs are included in the direct construction cost.

(gross) of electricity and achieved cumulative load, capacity, and availability factors of 0.64, 0.87, and 0.86 through June 1966.

Piqua. This organic-cooled and -moderated reactor plant achieved full power on Jan. 27, 1964. Facility operation through July 1966, during which many minor operational problems were encountered, produced 64,242,000 kw-hr of electrical energy. The availability and load factors for 1965 were 0.68 and 0.25, respectively.

Hallam. This sodium–graphite reactor achieved full power in July 1963. After operating for about 14 months, during which many problems were encountered, the plant was shut down for major reactor modifications.[9] Since then, a decision has been made to dismantle the facility. To the time the facility was shut down, 192,458,000 kw-hr (gross) of electricity was generated.

PROBLEMS

As of December 1965, except for the Carolinas-Virginia Tube Reactor, various problems prevented these six plants from becoming reliable electric generating stations during the initial operating period after achieving full power. However, the three boiling-water-reactor plants (Big Rock Point, Elk River, and Humboldt Bay) did achieve reliable operation after reactor or systems modifications were accomplished. Nearly all the 10 plants encountered discouraging schedule delays and additional costs and inconvenience even after the startup programs were completed.

Rod Drives. The most common problem encountered in the plants was the malfunction of control-rod drives. In general, the rod malfunctions were caused by either (1) unanticipated fouling from the reactor coolant or (2) unproved designs. Even after corrective measures were taken, the drives required periodic tests and inspection.

Steam Valves. In the facilities that employed a system to permit main steam flow to bypass the turbine and flow directly to the main condenser, the incidence of problems with the steam-bypass control valve was notable. Again, the root of the problem seemed to lie in the fact that the function is not conventional and that the designs are new. Above and beyond special problems encountered because the plants were nuclear, considerable problems arose with the conventional plant equipment.

GROUNDS FOR OPTIMISM

The 10 plants demonstrated that several different types of nuclear plants can be sited, constructed, licensed, and operated by small utility companies. Methods for doing so have been refined on the basis of the experience acquired with these plants. An important aspect of the projects was the acceptance by the public with only minor objections and few intervenors at public hearings for license applications.

These plants represented new technology, and some of the reactors successfully served as demonstration units leading to larger plants. Because of corrective action taken by designers as a result of the experience acquired with these plants, most of the specific problems and difficulties encountered with them will not be repeated in future plants. Other design improvements and standardization also have been derived from these plants. It should be noted that the costs (Table 3) of these early-technology plants are not representative of the particular type and size of plant and generally are considerably higher than the cost of a comparable small nuclear plant that might be ordered now.

Recognition of these factors is reflected in the fact that most of the utilities that participated in these 10 projects are seriously contemplating the installation of nuclear plants when they increase the generating capacities of their systems. (More than six of these utilities have already ordered new nuclear plants of larger sizes.)

Utility Market Potential

The interest in small plants raises questions as to the market potential for small plants and what the costs would be for second-round plants. The costs quoted for the 10 plants are not representative of the particular type and size of plant because most of these plants were early models, first-of-a-kind, or prototype plants. Thus their costs are considerably higher than the cost of an optimized plant that could be ordered today.

BY 1980: 15 NUCLEAR PLANTS OF 33 TO 66 Mw(e)

Both Southern Nuclear Engineering, Inc., and Kaiser Engineers evaluated the market potential for small nuclear power plants as part of the study. Southern Nuclear concluded that, between the years 1965 and 1980, there would be a market for ~300 steam electric-power units with a unit capacity of less than 100 Mw(e). The optimum size for a small nuclear power plant appeared to be either 33 or 66 Mw(e); for 33- or 66-Mw(e) plants, a potential market of 75 units was expected through 1980. Assuming a small nuclear plant could be developed which would be attractive enough to capture 20% of the potential market, they arrived at a figure of 15 plants which could be constructed.

BY 1972: 5 MORE NUCLEAR PLANTS AT 75 TO 125 Mw(e)

Kaiser Engineers surveyed the smaller utilities and considered all utilities whose load forecasts showed a need for installing a 75- to 125-Mw(e) unit

in any of the years between 1968 and 1972 to be potential candidates. Of the potential candidates, those cooperative or public-power utilities who paid gas prices of 30 cents/million Btu and coal prices of 27 cents/million Btu were identified as potential candidates for nuclear power. For investor-owned utilities, the breakeven prices were 43 cents/million Btu for gas and 37 cents/million Btu for coal. Based on these criteria, Kaiser identified 15 utilities with 23 plants in the 44- to 125-Mw(e) range and 10 utilities with 15 plants in the 75- to 125-Mw(e) range. As a result, they felt that there is a fair chance of being able to locate five small utilities where a nuclear power plant would be competitive.

HAZE IN THE CRYSTAL BALL

In view of the current lead time for nuclear systems, any market projections should be used with great caution. Also, load forecasts are subject to considerable change with the passage of time. The formation of pools, interchanges, etc., could change the picture.

Any future uses of small nuclear power plants will depend on whether small utilities are convinced that nuclear power plants will be the lowest-cost alternatives in the long run.

Small Plants Available

Reactor manufacturers were surveyed to obtain information for assessing the competitive economic performance of nuclear plants in small-power sizes. Kaiser Engineers also provided a general and economic assessment of small nuclear plants for use by small utilities.

A summary of the characteristics and projected costs of reactor types that reactor manufacturers feel are applicable to the present need for small civilian power plants is shown in Table 4.

POOL-TYPE REACTOR

The smallest plant offered was the DIPPER concept of the American Machine & Foundry Co. In this concept, electric-power costs would be reduced by sales of fresh water and isotopes as well as by taking advantage of the low-capital-cost features of a pool-type research reactor. Development costs were not included on the assumption that they would be written off on or before the first plant of this type.

SHIP-TYPE PWR'S

Compact shipboard reactor concepts may provide the utilities an opportunity to take advantage of reac-

Table 4 PROJECTED COSTS FOR PLANTS BEING OFFERED[2]

	Manufacturer							
	AMF*	UNC	UNC	GE†	GE†	GE†	AI	AI
No. of plants ordered	2nd	4	4	10	10	10	10	10
Power, Mw(e) net	2.5	16.5	51.6	50	75	100	50	100
Direct costs, 10^3								
Reactor plant		2,740	4,220					
Turbine plant		1,300	2,500					
Other		1,902	2,605					
Subtotal	2,867	5,942	9,325					
Indirect cost, 10^3	1,030	2,983	4,182					
Total capital cost, 10^3	3,897	8,924	13,507	11,558	14,867	17,780	11,330	17,870
Unit capital cost, $/kw(e)		540	262	231	198	178	227	179
Annual costs, 10^3								
Capital at 7%	195‡	625	945	809	1,041	1,199		
Nuclear insurance§	51	91	141	104	104	104		
Other capital charges	28	37	72					
Staff	115	316	363	196	209	218		
General & administrative	34	41	47	45	49	53		
Other operation & maintenance	159	90	123	122	146	168		
Fuel inventory	¶	79	176					
Fuel cycle	314	275	761					
Total, 10^3	896	1,554	2,619					
Energy costs, mills/kw-hr(e)								
Fixed charge		6.52	3.20	2.61	2.18	1.94	2.97	2.23
Operation & maintenance		3.86	1.45	1.04	0.71	0.63	1.67	1.16
Fuel		3.05	2.59	0.99	0.94	0.91	1.71	1.62
Total, mills/kw-hr(e)	15	13.43	7.24	4.64	3.83	3.48	6.35	5.01

*Includes isotopes and water production of 1 million gal/day at 80 cents/1000 gal.
†Derived from GE 10-year projections: 60% reduced fuel cost, 40% reduced staff, 33% reduced capital cost.
‡At 5%.
§Has not been calculated on a consistent basis.
¶Assumed waived.

tor designs with broader market potential. Development would be required to prove major component concepts. From the experiences with the small plants discussed earlier, the probability of an extended period of debugging would have to be assessed. The concept is offered by at least three manufacturers: Babcock & Wilcox Co., Combustion Engineering, Inc., and United Nuclear Corp.

ORGANIC REACTORS

The organic-cooled reactor offered by Atomics International is scaled up from the Piqua design and anticipates multiple orders and the design of more durable fuel. The economics and acceptance of the concept are contingent on development of a suitable fuel and on corrections of the problems experienced at the Piqua plant.

PWR'S AND BWR'S

The natural-circulation direct-cycle boiling-water reactor (BWR) and the pressurized-water reactor (PWR) are the only commercially developed plants that could be purchased in the small sizes. In sizes above 50 Mw(e), and under certain conditions, it appears that the BWR offers near-term competitive application for many utilities. The concept would be an improved version of the Humboldt Bay design. The cost reductions projected by General Electric have the following stipulations:

1. Multiple orders (5 to 10) of duplicate plants for each vendor could reduce capital cost 12 to 15%. The improvement would be achieved primarily by using a single architect-engineer for all plants and spreading plant design costs over all plants; it also includes 3 to 5% lower equipment and service costs due to multiple purchasing of equipment components. The gain is divided evenly between the nuclear boiler and the balance of the plant. The estimate allows for adjustments in condenser-coolant intake and discharge facilities and foundation requirements for the various sites.

2. Major simplification of plant design could reduce capital cost 10 to 15%. This would include safety features using proven concepts but fewer systems, each of which would be highly reliable.

3. Location of the nuclear plant on an existing site of other thermal plants could reduce capital cost 5 to 10% by taking advantage of common support facilities.

4. Nuclear fuel improvements are forecast to reduce fuel cost 4 to 6%/year for the next 10 years.

5. Reduction in number of plant personnel by 25 to 40% would save $50,000 to $100,000/year. This gain would be permitted by simplified plant design and from the utility's contracting for maintenance and technical services only as required.

6. A 3- to 6-month shortening of the licensing process, with comparably shortened construction schedules, could reduce capital cost by 1 to 3%. This shortening would be possible if the AEC and other regulatory organizations accept proposed standard designs for local siting and licensing without requiring extensive investigation of each application.

ADVANCED CONVERTERS AND BREEDERS

Table 5 presents characteristics of advanced converter and breeder reactor designs submitted by Westinghouse for future small nuclear power plants.

Table 5 FUTURE SMALL POWER REACTORS[2]

	Fast Breeder	Advanced PWR
Power, Mw(e)	75	100
Mw(t)	230	265
Coolant	Heavy water	Light water
Pressure, psia	3400	3400
Temperature, °F	720	960
Breeding ratio	>1.2	
Median fission energy, kev	25	
Core height, in.	57	72
Diameter, in.	36	56
Fuel-pin diameter, in.	0.30	0.30
No. of subassemblies	37	457
Vessel diameter, ft	7	7
Height, ft	15	16.7
Wall thickness, in.	7/8	7/8
Steam pressure, psia	1200	3400
Temperature, °F	640	960
Cost	25 to 30% reduction in capital cost under current PWR costs	

Westinghouse indicated the concepts offer a potential for reducing plant cost 25 to 35% in addition to the breeding gain, which would reduce fuel-cycle costs. Although the plant concepts are very attractive, considerable development would have to be done on these plants.

Economic Status

Tables 6 and 7 show the results of Kaiser Engineers' economic assessment of capital and energy costs now and for future reductions. Costs for the pressurized-water reactors probably would be similar to those for boiling-water reactors because, if an actual project were being considered, the manufacturers of boiling-water reactors and pressurized-water reactors likely would bid competitively.

Assumptions. The reduced costs were developed assuming that joint action of five utilities takes place and that standard plants are ordered, that joint licensing and safety studies take place, that fuel is purchased jointly, and that joint action takes place during operation to reduce operating costs. Reduced fuel-cycle costs were also anticipated.

Comparison. Figure 1 shows a comparison of estimated power costs as a function of plant size. Also shown are the results of the utility survey showing present power costs as a function of peak load. The

Table 6 PRESENT CAPITAL AND ENERGY COSTS[3]

Cost item	Type of reactor				
	PWR-CNSG	OCMR		BWR	
	25 Mw(e)	50 Mw(e)	100 Mw(e)	50 Mw(e)	100 Mw(e)
Capital costs, 10^3					
Total without fuel	15,000	19,000	25,000	19,500	28,000
Nondepreciated capital for fuel	1,350	2,320	4,450	3,360	6,290
Total capital required	16,350	21,320	29,450	22,860	34,290
Unit energy costs, mills/kw-hr*					
Total unit cost at 60% P.F.†	17.4	12.1	8.6	12.4	9.0
Total unit cost at 80% P.F.	13.8	9.8	7.1	10.2	7.6

*Average for first 10 years of plant operation.
†P.F. = plant factor.

Table 7 REDUCED CAPITAL AND ENERGY COSTS[3]

Cost item	Type of reactor				
	PWR-CNSG	OCMR		BWR	
	25 Mw(e)	50 Mw(e)	100 Mw(e)	50 Mw(e)	100 Mw(e)
Capital costs, 10^3					
Total without fuel	12,600	15,950	21,800	16,930	25,150
Nondepreciated capital for fuel	1,230	1,840	3,490	2,350	5,000
Total capital required	13,830	17,790	25,290	19,280	30,150
Unit energy costs, mills/kw-hr*					
Total unit cost at 60% P.F.	14.0	9.5	6.9	10.0	7.6
Total unit cost at 80% P.F.	11.1	7.7	5.7	8.2	6.4

*Average for first 10 years of plant operation.

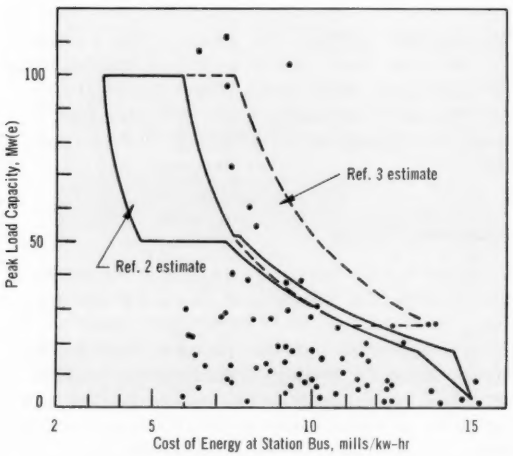

Fig. 1 Power cost as a function of plant size, based on two estimates and utility cost data.

power costs indicated as Ref. 2 estimates are the results of the manufacturers' estimates, and the Ref. 3 estimates are Kaiser Engineers' economic assessment of both present and future energy costs with varying plant-capacity factors, with the lower limit of costs for an 80% plant-capacity factor and the reduced costs previously discussed.

Conclusions

The general conclusions that can be drawn from the study are:

1. Experience has demonstrated that several types of small nuclear plants can be sited, constructed, licensed, and operated by utility companies.

2. Present nuclear power systems on a one-of-a-kind basis are only marginally competitive in most areas of the United States.

3. There is a possibility of significant cost reductions in small nuclear plants through joint action; this would make nuclear power in unit sizes over 75 Mw(e) competitive in some areas of the United States.

I want to thank Richard J. Gariboldi of the Chicago Operations Office of the U. S. Atomic Energy Commission for helpful discussions of the study on which this article is based.

References

1. S. F. Burton and A. G. Hosler (Eds.), Small Nuclear Power Plants. Vol. 1. Design, Construction, and Operating Experience, USAEC Report COO-284(Vol. 1), Chicago Operations Office, October 1966.
2. Chicago Operations Office, Small Nuclear Power Plants. Vol. 2. The Industrial Expression of Supply and Demand Considerations, USAEC Report COO-284(Vol. 2), March 1967.

3. Chicago Operations Office, Small Nuclear Power Plants. Vol. 3. A General and Economic Assessment, USAEC Report COO-284(Vol. 3), March 1967.

4. B. A. Schmeh, Operational Data for Power Reactors in the United States, *Power Reactor Technol. and Reactor Fuel Process.*, 10(1): 32-58 (Winter 1966-1967).

5. G. F. Bierman and W. J. Miller, Nuclear Plant Performance—Good and Getting Better, *Power Reactor Technol.*, 9(3): 110-122 (Summer 1966).

6. F. H. Martens, Design Foresight Can Simplify Reactor Modification, *Power Reactor Technol.*, 9(2): 74-79 (Spring 1966).

7. R. C. Skaardal, Operational Problems and Solutions, *Power Reactor Technol.*, 8(4): 271-279 (Fall 1965).

8. E. A. Wimunc, How Serious Are Vessel Cladding Failures? *Power Reactor Technol.*, 9(3): 101-109 (Summer 1966).

9. F. A. Smith, Hallam Operation in Retrospect, *Power Reactor Technol.*, 8(4): 279-289 (Fall 1965).

Burnup Predictions for Thermal Reactors: A Synthesis of Approximations

By Bernard I. Spinrad*

Perhaps the most formidable problem of reactor physics is burnup prediction that calls for solutions for all the classical problems of reactor theory. In practice, approximations are necessary. After examining the goals of burnup physics, this article reviews basic burnup theory, discusses the general theories used for survey purposes and the more precise theories used in design, and then considers specific applications. You may conclude, as I do, that our theories have become overly elaborate.

Goals of Burnup Physics

As explained in the subsection "Diversity, a Hallmark of the U. S. Reactor Program," the differentiation of methods used in burnup-physics studies is largely a matter of organization. The three separate goals of burnup physics also distinguish classes of methods.

To Aid Design. Once the basic reactor concept has been decided upon, there remains the question of how to deal with the fact that fuel burnup alters the reactor. In this sense, burnup is primarily a reactor, rather than a fuel-cycle, problem. The computations wanted are those which answer the following kinds of questions:

- How much excess reactivity should be loaded in initially?
- How much control should be provided for burnup?
- As burnup proceeds, do the engineering limits change? Does the point of closest approach to burnout shift? Does its level shift? Does the flux shift enough to change the prediction of irradiation damage to the vessel?
- Does the pattern of rod movements required to trim reactivity lead to hot spots? Should burnable or soluble (where appropriate) poison be used?
- Do the reactivity coefficients change enough so that trimming of the parameters of the dynamic regulating system should be permitted? How little trimming can we get away with?

Because the design problems arise from quite detailed considerations, answers to the problems require, primarily, that the theory be precise.

To Expedite Operation. Although a careful design is based on and directs a fairly detailed operating regime, nevertheless, inspection tolerances, component performance tolerances, and the desire to override small mistakes in both prediction and operation lead to some operational option. The operator's questions are generally strategic or dynamic:

- If I disturb the symmetry of control-rod placement to counteract an observed power asymmetry, will this lead to increased power-flattening difficulties later?
- Should I be concerned if the operational power coefficient is not the same as predicted? Is it likely to depart further?
- Is my unaccounted reactivity change (according to the rods) worth worrying about? Should I recalibrate the rods? Recommend a change in fuel scheduling?
- One of the monitors is out. Can I get along without it?

The operator has experimental data. Although his questions are qualitative, they are pressing. As an operational aid a predictive computational system is needed, either on- or off-line. The system must provide fast, qualitatively correct answers, but they need not be very precise. We expect that the system will be used frequently. Therefore the appropriate theoretical scheme must be simple, fast, and inexpensive; it should be highly modular because so many of the problems are detailed questions. Ideally, it should include several alternative methods of attack because no single scheme is likely to be appropriate for all problems.

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Diversity, a Hallmark of the U. S. Reactor Program

Central-station nuclear power in the United States benefits from a great diversity of effort. Substantial programs have studied many types of reactors: D_2O-moderated lattice reactors with various coolants, spectral-shift pressurized-water reactors, uniform-lattice PWR's, seed-blanket PWR's, boiling-water reactors, graphite-moderated lattice reactors with compact fuel and various coolants, graphite-moderated reactors with dilute fuel, aqueous-, molten-metal-, molten-salt-, and other fluid-fuel reactors, steam-cooled thermal reactors, and fast reactors.

THE EMERGENCE OF STYLES

Each company in the reactor business in the United States has recognized a commercial requirement to develop its own reactor types, modifications, and style of investigation. Each company differs in terms of facilities, personnel, and judgments as to what are important unknowns. The various national laboratories also have their individual styles. Thus the U. S. reactor program is best described as a synthesis of programs in numerous places, with most of the programs having evolved locally to meet objectives and circumstances at each place. Coordination by the AEC prevents unnecessary duplication of effort and assures that programmatic "blind spots" do not develop; the American Nuclear Society (ANS) encourages and advances professional competence and knowledge.

THE SUBTLETIES OF THEORY

The institutional arrangements of AEC and ANS attempt to make all theoretical tools available to all who might need them. However, it is apparent that the transfer of programs from one computing device to another is not a trivial matter. A number of codes use very similar theories and, in principle, the same data; many of the codes were set up because it was easier to program anew than to debug a borrowed program. However, in so doing, the subtle shifts of emphasis from one installation to another contribute real differences in code performance. Thus, before translating a code, a code borrower would do well to study the background design reports and theoretical papers of the code originator: the organizational style is generally reflected in the code.

For all these reasons the standard characterization of reactor theories according to reactor type is now not very significant in the United States. Except for BWR's and highly enriched seed-blanket PWR's, which require special techniques, the preferences of the design groups are the significant classification parameters.

Predictive burnup theories appropriate for use as operational aids are in early stages of development. They are strongly related to the topic of computer-aided operation, which is likewise not a mature subject. Therefore this article will not treat the subject explicitly.

To Plan Fuel-Cycle Program and Extend Design Concepts. For these, general properties of fuel cycles are desirable so they can be compared. Therefore the appropriate theoretical models lack detail in explicit reactor description; in fact, a model that reduces the reactor to a black box represented by a few parameters is preferred.

Basic Burnup Theory

The equations of isotope formation and destruction in a reactor are intrinsically simple: nuclides are formed by neutron capture in a predecessor, by radioactive decay, or as products of fission; they decay by neutron capture or fission or by radioactive decay. The differential equations describing the changes in isotope concentrations with time are as functions of concentration, linear and first order in time. The solution of these equations is a straightforward mathematical process; it is almost trivial on a modern computer, even for a large set of nuclides to be considered.

VARIATIONS IN SPACE AND TIME

The complicating factor on this simple picture is the fact that the coefficients of the concentration terms in the isotope equations can vary in space and time. This is so because the processes that depend on neutron capture and fission occur at rates proportional, not only to material concentrations, but also to neutron flux and effective cross section.

Neutron Intensity. Neutron intensity is both a time- and a space-dependent variable. As a time-dependent variable, it may be removed from consideration by replacing intensity and time with some measure of flux time such as fission-heat production; but even this simplification is an approximation which fails when (as with xenon-transient or ^{233}Pa-concentration calculations) neutron capture and radioactive decay processes are both significant. It is nevertheless conventional, and correct, to make this simplification for power reactors, whose isotopic changes are dominated by neutronic processes. However, this leaves a residual dependence on space, through flux being a space variable. At best the burnup problem requires a large bookkeeping operation to follow the nuclides in time over all the points of the reactor volume.

Neutron Spectrum. Neutron spectrum enters into the definition of cross section, the other factor of the coefficients in the isotopic-change equations. If we normalize flux time to some measure of fission heat, we implicitly normalize all cross sections to the effective value for one (or a few) species over the neutron spectrum. However, each neutronic process is characterized by its own spectral dependence of cross section. For the flux-time definition adopted, this implies an effective cross section that varies if

the neutron spectrum varies; and, indeed, the spectrum will vary over space and time.

REACTIVITY CHANGE

Isotope evolution rates vary from point to point because of spatial variation of flux. Even if the spectra in a reactor were originally the same everywhere, the changed evolution rates would lead to varying point-to-point spectra; and changes in both spectra and isotopic concentrations lead to shifts in spatial flux and spectrum with time.

Control. The situation is made more complex by the changes in isotopic concentration, neutron spectrum, and flux shape because these changes lead to reactivity changes. To compensate requires a reactivity shim system. Therefore a real reactor does not burn up in a determinate way. The design and operational tactics of the control system give an additional degree of freedom that is required to keep the reactor running.

Refueling Strategy. A further operational degree of freedom is provided by the refueling strategy. Since the substitution of fresh fuel for burned fuel compensates reactivity loss, it is possible, by frequent refueling, to maintain a reactor in a pseudostatic state without creating major demands on the control system for shimming. It is also possible, by less frequent refueling, to achieve compromises between excessive control requirements and undesirable shutdowns.

NECESSARY APPROXIMATIONS

Thus, despite the apparent mathematical simplicity of the equations, the burnup problem is perhaps the most formidable one of reactor physics. It requires solution of all the classical problems of reactor theory, as subroutines. To solve a complete burnup problem, one would have to do so "exactly" by solving a complete reactor system, with nonuniform material concentrations, spectra, and control elements for each of a large number of time steps. Each change in control tactics or fueling strategy would require another such set of problems. As a practical matter, this procedure is impossible. Therefore burnup theory is forced to become a synthesis of approximation methods. The goal of these methods is to reduce the number of detailed total reactor calculations without unduly sacrificing precision.

Simplifying Assumptions. Three general categories of reduction are used: (1) the computation of idealized reactors and fuel cycles, such that reactor properties are fixed and only isotope evolution need be followed, (2) reduction of the amount of detail used in modeling the reactor in time and space, generally to interpolate results between well-computed points, and (3) the use of correlations from previous results to reduce the number of variables that must be considered or to furnish good approximations for tactical and strategic purposes. All these methods require calibration against a number of detailed reactor computations. Comparison with experience is of value primarily to correct input data, such as cross sections. The reason is that general experience provides only integral checks, which shed no light on such matters as error cancellation; also, experience is commonly less precise than theory. However, once a reactor has been operated, the specific experience can be formulated into reliable correlations.

Theories for Survey Purposes

Although generally categorized as zero dimensional, the more significant assumptions of theories used for survey purposes concern spectrum. If the spectrum of a region can be taken as a property of the region rather than of the whole reactor, great simplifications are possible. In particular, the cross sections of the isotopes can be procured simply as functions of isotopic concentrations, without reference to total reactor flux. If the separability of spectrum is a valid approximation, neutron flux can be calculated without any iteration for regional cross sections, and the equations of isotopic change can be integrated directly. Even if the spectrum-separability approximation is not quite valid for detailed design, its approximate validity simplifies survey work.

CONSTANT SPECTRUM

Separability of spectrum is itself a significant, potentially useful approximation. An even simpler approximation is that of constant spectrum; it is from this approximation that most early burnup physics derived.[1,2]

Constancy of spectrum is not an absurd situation; in some cases it may be closely approached physically. For example, the uniform-graded-irradiation scheme[1] is one of constant spectrum that is a limiting case to a refueling scheme in which fresh fuel is frequently added in small increments to replace burned fuel. Also, bidirectional incremental loading[3] of fuel slugs in large reactors is reasonably approximated by a constant-spectrum model.

To some extent, spectrum changes that are not small can be washed out in survey calculations. First of all, most of the cross sections of heavy elements have crudely similar characteristics; approximately $1/v$ dependence, similar resonance structures, and so on. Thus spectral changes are reflected in cross-section changes that are relatively smaller. Additionally, the use of macroscopic cross section, rather than isotopic abundance, as the dependent variable[1] suppresses the effect of variation of microscopic cross section on the results.

Finally, it has been pointed out[4] that experimental results can be correlated well by a constant-cross-

section theory. At the panel where these data were presented, the generally accepted explanation was that the greatest errors in effective cross section occur during early stages of buildup on an isotope, when details of resonance self-shielding are important. However, these errors do not contribute much to the error in concentration of the isotope after it has built in to a significant degree and become "thick" to its own resonances.

Under the constant-spectrum approximation, a set of microscopic cross sections is invented and used for a series of calculations. Initial concentrations of materials may be varied and leakage effects may be assessed by use of a DB^2 term. Control can be simulated in various ways because, in such a model, "control" is merely excess absorption to be added to make the system critical. Because the reaction rates of all the isotopes can then be obtained by simple quadrature, a large quantity of data can be obtained to give reactivity, production, and conversion of heavy isotopes. These data are particularly useful in surveying control tactics and fuel management; they give the "grand picture" in a way that is qualitatively correct.

With modern computers the programs to solve the burnup equations in this constant-spectrum approximation are trivial. The lack of recent literature on this subject does not imply that the approximation is not used; it is just that it is no longer appropriate to document such use, which is widespread.

LOCAL SPECTRUM

A much better approximation than that of constant spectrum is that the spectrum is separable from the spatial dependence. After all, the spectrum describes neutrons born within one migration length of the point under investigation, whereas typical power reactors share reactivity over the much larger distances characterized by $1/B_m$ (B_m^2 is the material buckling). Even near reflectors the major effect on cross sections frequently arises from shifts in thermal and near-thermal spectra, whose equilibration distance is smaller than L, the thermal diffusion length.

Thus qualitative reasoning suggests that this approximation should be quite good; indeed, it forms the basis of many computational schemes for both survey and design purposes.

If the spectrum is not explicitly dependent on the flux shape, it follows that the effective cross section of any given nuclide is a function of the concentrations of the nuclides present in the region. Leakage may be incorporated by way of a buckling term; however, if the dependence of the cross sections on buckling is nontrivial, the approximation will clearly fail. Simplified computation schemes may be recovered by using a group structure for the spectrum, for which the contributions of most groups to effective cross section are essentially independent of the buckling; but this is really a different approximation.

The dependence of effective cross sections on nuclide concentration can be worked out from the basic nuclear data in each case. However, this is not really necessary; a logical result of the approximation is that, for any given initial fuel, the evolution of isotope concentrations is a function only of flux time, or irradiation. Thus the solution of the complete, zero-dimensional system, done for each time step, produces a result that can be used forever after.

The solution may be retained in any one of a number of ways, such as: (1) a matrix of isotope concentrations for various points of flux time, (2) a matrix of parameters of fitting functions for these concentrations, and (3) a matrix of effective cross sections, or fitting functions. The preferred form for retention depends on the computing device and the relative speed of memory search, interpolation, and computation. The storage of cross sections rather than concentrations is indicated when cross-section variation is small and easily represented, when computation is very fast compared to memory retrieval, or when the results from a range of starting materials are to be represented by formulas or interpolation tables.

SPECIFIC CASES

Breen[5,6] has presented a scheme for representation of group cross sections as functions of nuclide concentrations (HARMONY). Although this scheme is intended for use only within groups of a multigroup system, its use in a single-group representation is straightforward.

Revised GAD is a typical program for computing fuel changes by a multigroup procedure.[7] A spectrum is computed from the macroscopic multigroup cross sections, fission spectrum, and inserted slowing-down (and -up) cross sections. This spectrum is then used to determine effective cross sections for a burnup step. After the burnup step the new cross sections are used to compute a new spectrum, and so on. Leakage may be included as DB^2, and a number of reactivity checks and fueling options are included for survey calculations.

ALCHEMY[8] and MELEAGER[8,9] are two codes in use at Hanford for more or less precise point-spectrum burnup calculations.

Other reports in the literature (see the bibliography of Ref. 2) illustrate a variety of similar codes. The ones cited here are not to be taken as necessarily the best, but simply as typical.

It must be pointed out that the "local spectrum" approximation is, for lattice reactors, an approximation. Common practice is to compute a single cell as a "local spectrum" point, introducing fission neutrons and considering fast effect, intracell neutron migration, resonance absorption, thermal migration, etc. In addition to mapping out isotopic evolution, such a calculation also furnishes local k_∞ and B^2, which are also retained for inspection.

How Much Sophistication? Many of the modern codes use the most sophisticated spectral and cell disadvantage theories available. I believe, in spite of the weight of opinion to the contrary, that this degree of refinement is generally a waste of time. The effective microscopic cross sections that result ought to be available from simpler theoretical calculations to which small correction factors can be attached; the utility claimed for HARMONY[6] seems to bear out this contention.

Reactor Design Theories

As opposed to survey theories, design theories are expected to be more detailed and precise. The spatial independence of spectrum is a useful simplification, but is not considered adequate. However, it does point the way to a commonly used representation which, by its success, is justified.

The representation is a few-group one. Within each group the spectrum may be calculated or estimated as a concentration-dependent function, but one which does not depend on flux shape or total spectrum. Thus, given a set of isotope concentrations, local, coarse-group cross sections may be computed. From these a few-group calculation for the reactor can be made, yielding group fluxes as a function of position. Then these group fluxes can be used to weight the group cross sections at each point, yielding effective reaction rates for the burnup step. Then the computations are iterated.

GROUP-SPECTRUM SOLUTIONS

The burnup steps in the system just described are the simplest part of the calculation, bookkeeping problems aside. On the other hand, solution of the intragroup-spectrum problem is tedious, particularly if a large number of points are to be covered by cell calculation. Generally, therefore, intragroup spectra are held constant over fairly large burnup intervals. As a refinement, effective cross sections may be interpolated between these intervals.

The computation of intragroup spectrum (within which category, for the purposes of this article, disadvantage factor calculations are included) may be made by any of a large variety of design methods and codes, according to the attitudes and prejudices of the designer. Westcott factors, THERMOS, MUFT, SOFOCATE, fine group S_n, etc., have all been used.

With experience, a mathematical analysis of results from a number of sample calculations ought to be available as correlated microscopic cross sections, such as those exhibited in the HARMONY library. Apparently a most useful preparatory program for burnup work would be one that takes detailed calculation results and performs as much data compression as possible. I know of no such general program, but I consider it to be a useful new item of business.

"WHOLE-REACTOR" SOLUTIONS

The European–American Committee for Reactor Physics (EACRP) has taken the position that the actual flux calculation over the reactor is the most troublesome problem in burnup theory. I agree. Even with reduction of the total spectrum to a few groups, the reactor is nonuniform; after a burnup step, its properties vary continuously from point to point.

One point is worth emphasizing: there are several things that are estimated from the flux calculation, but they are not necessarily computed properly by the same methods. For burnup calculations the important result is the spatial distribution of neutron reactions; for this, computed flux must be a good approximation to actual flux. An attempt to specify variation of reactivity parameters, such as changing control-rod calibrations, dynamics, or global reactivity coefficients, does not require as good a flux specification to get high precision.

The significance of this point is that a large number of approximate methods for calculating flux are founded on a variational principle. Unless the parameters to be minimized are significant parameters of the flux shape, these approximations must bear the burden of proof that they do, indeed, give a correct spatial distribution.

CASES AND COMMENTS

For examples of the various computing systems that include burnup for design purposes, the reader is referred to the references. Design systems must include, at least: (1) a spectrum of macroscopic-cross-section module, (2) a burnup module, (3) a reactivity-change module, (4) a control-option module, and (5) a flux-calculation module. Since there are valid reasons for including several alternative modules of a given type, the concept of a burnup code probably must be discarded in favor of a system of linked modules and paths, as in the Argonne Reactor Computation System, ARC.[10]

Specific Applications

Of the many reactor types that have been studied in the United States, three types of reactors that are cooled and moderated by light water have been studied most extensively. These are the seed-blanket pressurized-water reactor (PWR) (as Shippingport[11]), the uniform-lattice PWR (as Yankee[12]), and the boiling-water reactor (BWR) (as Dresden[13]).

SEED-BLANKET PWR's

The Shippingport reactor type has both its reactivity and power shape dominated by the properties of the enriched seed, in which are also embedded the major control rods. The seed is a thin region of fast neutrons, only a few root ages across, and is highly absorbing to thermal neutrons. Most of the neutrons

absorbed as thermal are captured in a plate quite near the water channel in which thermalization occurs. The seed also contains a significant amount of burnable poison that is precisely located and lumped so as to present a minimum of reactivity change and power shift during burnup.

Power Density. Since a seed-blanket system must have a highly rated seed for the blanket to have a significant power density, burnout heat-transfer margins are trimmed to a minimum and must not be exceeded during seed life. Nevertheless, there is much local shifting of power according to control-rod motion and because of the large depletion desirable in a seed. Both for design and for operation analysis, precise determination of power density is necessary.

The closest analogy to these requirements in other reactors is found in design studies of high-flux reactors such as HFIR[14] and AARR.[15]

Study Sequence. The scheme used for burnup studies can be briefly described as follows:[16]

First, the whole reactor is represented as a detailed, three-dimensional, multigroup system. Relatively homogeneous regions, such as the blanket and the more uniform portions of the seed, are represented as homogeneous regions. (The degree of homogenizing might be varied from group to group to save computing, but this has not been done in practice.)

A limited number of different reactor situations (a couple of different burnups, a few control-rod settings) are computed in full detail. Primarily, the results of these computations are used to check methods.

A few-group model is achieved by noting that local spectra are determined mostly by local conditions over limited ranges of energy. This is the HARMONY system. The true spectrum at a point is represented by an N-group discontinuous spectrum, in which discontinuities at energy breakpoints may exist.

A fewer region model is achieved primarily by physical intuition, aided by checking synthesis calculations against the 3D check points. The core is examined, layer by layer, for variations in material composition and configuration. Layers of different properties have their fluxes computed by a 2D code, such as PDQ. Then the layers are separated into areas, within each of which the planar flux shape is taken as a trial function for the next stage of calculation.

Then a detailed calculation is performed on the fresh reactor in the few-group model. The flux calculation is by variational synthesis.[17,18] A burnup step is then taken, isotope inventories altered in detail, new group-region cross sections computed, and a new variational flux calculation is made. This procedure is iterated throughout the core life. Occasionally a more detailed few-group 3D relaxation is interspersed to see if the flux shapes in all the major regions continue to be reasonably well approximated by the variational trial functions used. When this does not seem to be the case, a new set of 2D calculations is performed.

After a few runs of this sort, there is less need for repeating the 2D calculations. A library of trial functions, once achieved, is retained, and the proper set of functions to use at any step can be programmed.

Results. The results of this procedure have been excellent. The two major reactor limits are (1) burnout for flux shape and (2) seed life at the time of total withdrawal of control rods. In the former case, best calculation methods predict "hot spot" magnitude correctly to well within 10% in special critical experiments. (Here, and elsewhere, flux-shape errors correspond crudely to observed theory–experiment discrepancies between power-density maximum to average, these averages being taken over only the higher flux regions of the core.) In the latter case, seed-life predictions have accuracies which imply that reactivity change is about as well known as for other PWR's. This precision is achieved, of course, with the help of a normalizer obtained from beginning-of-life experience.

The high precision achieved here must not be considered to be a demonstration of universal applicability of the methods used. As previously mentioned, the approximations have only been tested against seed-blanket burnable-poison systems.

Computing times and costs for burnup calculations by these methods, although not trivial, are not an inordinate fraction of reactor design costs.

LATTICE PWR's

The techniques used for seed-blanket systems have been extended only to other enriched-reactor designs. On the other hand, those used for lattice PWR's are typical of techniques for other lattices as well. Most have been described by Sofer et al.[19]

The fundamental tools are: (1) a multigroup treatment of slowing down (of the MUFT type), (2) a representation of fast effect and resonance capture within this structure by use of group cross sections derived from lattice-theory calculations, and (3) the inclusion of ^{235}U and plutonium cross sections by a combination lattice–thermalization-law method such as THERMOS or SOFOCATE.

Such effects as may be caused by nonuniform burnup and buildup of isotopes within a fuel rod are estimated by ancillary calculations, whose results are used, where necessary, as burnup-dependent corrections to the appropriate cross sections.

Reactor flux variation is generally a very smooth function in such reactors, control rods excepted; thus the spatial representation is normally in a coarse mesh, and control calculations are included as a superposed sink.

For the spatial calculation, the multigroup representation is generally collapsed to about four groups. The finer group structure is used almost exclusively to obtain group transfer and reaction cross sections for the coarse groups.

Because of the relatively smooth variation of flux, the burnup time steps usually can be generous. If too large a step has been taken, the computation may be salvaged by interpolation. (The judgment of "too large" depends simply on flux shape because several burnup steps are computed between flux recalculations.)

Results. The results of such computations have been compared with experimental data from the Yankee core.[20,21] Agreement has generally been excellent on flux shape, reasonably good on reactivity, burnup, and isotope production and again excellent when isotope production is compared against burnup. Some disagreements of prediction against experimental changes in reactivity coefficients have been noted, but their origin is obscure and may not be neutronic.

Code systems used for this work are named LEOPARD[22] and LASER;[23] they differ from each other in the thermalization model used.

BOILING-WATER REACTORS

Although commonly loaded in a lattice arrangement, boiling-water reactors differ in two significant and fundamental ways from all others. First of all there is a strong reactivity and flux effect due to boiling; this is manifested as a change of reactor properties along the coolant flow path. Second, as a corollary, the coolant flow direction defines an axial direction for the reactor as a whole, far more strongly than is done by fuel orientation.

A third difference from other reactors actually serves as a simplifying principle to the reactor physicist: the reactivity is strongly affected by void concentration along the axis; but hydrodynamicists can only specify void concentration to limited accuracy. As a result it is logical to believe the physics reactivity calculation and to adjust to criticality by varying a hydraulic parameter, such as a steam–water slip-velocity ratio.

Vitiating all these observations, but not eradicating them, is the fact that most boiling reactors are designed to have not too large a reactivity gradient between top and bottom under operating conditions.

Until now the lattice and enrichment selection has been of cores that are slightly overmoderated at full water density and slightly undermoderated at exit-void density. Such a system has a strong negative power coefficient if operating power is significantly increased, but a milder one in the operating range.

Simplified Neutronics. The extra hydraulic complexity of BWR's makes a simplified neutronic model even more necessary as a practical matter. For this purpose a local-spectrum approximation has always been used. In estimating the local spectrum, the most significant variable is water density.

As with pressurized-water reactors, the basic tool is the "cell" calculation that is performed to yield neutron-reaction rates. However, BWR cell calculations are performed at several different water densities. In addition to reaction rates, these calculations yield one-group values of B^2, Σ_a, and D.

An iterative procedure is then carried out to determine the power shape. The first step involves assuming a power shape, which yields a void distribution that defines the one-group lattice parameters spatially for neutronic calculation. Hydraulic considerations are such that the axial void distribution has only limited freedom compared with the radial (entrance voids are zero, and voids increase along the channel; exit voids are governed primarily by total channel power). The neutronic calculation yields a reactivity for the system that implies a critical setting of rods or poison. It also improves the eventual power shape.

Given power shape, a point-by-point depletion calculation is then performed and the sequence of calculations repeated.

The actual power-shape, reactivity calculation can be done in several ways. Reasonable success has been achieved at Argonne with 1 by 1 dimensional calculations.[24] The estimates of void are used to determine what amounts to a radial buckling for each radial region of the reactor; the radial power shape is solved as a 1D or 2D problem. There has been little need for iteration. At Argonne total reactivity errors in the theory are generally ignored in favor of adjusting hydraulic conditions as a parameter. Predictions of reactivity have always been as changes relative to initial conditions.

The FLARE[25] program has been pioneered by General Electric Company. This program includes as its neutronic step a one-group 3D coupled-reactor-region calculation. FLARE also incorporates the iterations already described.

Results. Whole-reactor reactivity is claimed to be, on the whole, well predicted (within 2%). By allowing an eigenvalue normalization at the beginning of life, or in some other clean configuration, this error is cut in half. Power shape seems to be predicted within 10%.

A Conclusion

The fact that these very simple codes for neutronic calculation of boiling reactors appear to do as well as the more elaborate ones used for PWR's is the main justification for my contention that our theories have perhaps become overly elaborate. Although it is true that these simpler calculations require eigen-

value normalization, the eigenvalue (total reactivity) is always directly observable and thus is a most reliable input to any predictive tool.

This article is based on my paper, "Fuel Burnup Predictions in Thermal Reactors: A Review of Recent Work in the U.S.A." presented to the IAEA Panel on Fuel Burn Up Predictions in Thermal Reactors, Vienna, Apr. 10–14, 1967. The coverage of burnup theory here is very brief. For further background information and bibliographies, the reader is encouraged to consult the references, particularly Refs. 19 and 26, which present an excellent background of U. S. work in the burnup field; Ref. 27, which contains a great deal of information of continuing value; and Refs. 28 and 29, as well as WCAP reports with numbers between 6050 and 6080, for extensive coverage of the Yankee program; and other reports of the WCAP-3269 series, for earlier design methods used in PWR's.

References

1. B. I. Spinrad, J. C. Carter, and C. Eggler, Reactivity Changes and Reactivity Lifetimes of Fixed-Fuel Elements in Thermal Reactors, in *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955*, Vol. 5, pp. 125-144, United Nations, New York, 1956.
2. W. B. Lewis, Some Economic Aspects of Nuclear Fuel Cycles, in *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955*, Vol. 3, pp. 3-13, United Nations, New York, 1956.
3. W. B. Lewis, Low Cost Fuelling Without Recycling, Canadian Report AECL-382, Dec. 10, 1956.
4. B. H. Duane, Pacific Northwest Laboratory, Synthesis of Transmutation Codes and Theory, summary presented to panel on Reactor Synthesis Computer Codes for Burnup Analysis, ANL, January 1967.
5. R. J. Breen, The Representation of Few-Group Cross Sections in Multiregion Diffusion–Depletion Calculations, *Trans. Am. Nucl. Soc.*, 7(2): 292 (November 1964).
6. R. J. Breen, O. J. Marlowe, and C. J. Pfeifer, A System for Nuclear Reactor Depletion Computation, in Proceedings of the Conference on the Application of Computing Methods to Reactor Problems, USAEC Report ANL-7050, pp. 265-278, Argonne National Laboratory, August 1965.
7. F. W. Todt, Revised GAD, An Infinite Medium Depletion Program for Reactor Fuel Cycle Analysis, USAEC Report GA-6635, General Atomic Division, General Dynamics Corp., Nov. 10, 1965.
8. B. H. Duane, Transmutation Logic for S_N Transport Analysis, in Proceedings of the Conference on the Application of Computing Methods to Reactor Problems, USAEC Report ANL-7050, pp. 291-298, Argonne National Laboratory, August 1965.
9. E. A. Eschbach, D. E. Deonigi, E. T. Merrill, and W. I. Neef, A System of Chained Computer Codes for Reactor Fuel Cycle Analysis, in Proceedings of the Conference on the Application of Computing Methods to Reactor Problems, USAEC Report ANL-7050, pp. 327-345, Argonne National Laboratory, May 17, 1965.
10. C. N. Kelber et al., The Argonne Reactor Computation System, ARC, paper presented at International Conference on Research Reactor Utilization and Reactor Mathematics, arranged jointly by the Reactor Group of Centro Nuclear de Mexico and the Mathematics and Computation Division of the American Nuclear Society, Mexico City, May 2–4, 1967.
11. P. G. DeHuff, W. R. Ellis, J. C. Grigg, R. V. Laney, J. C. Rengel, and M. Shaw, Experiences in the Design, Construction, and Operation of the PWR at Shippingport, in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*, Vol. 8, pp. 47-85, United Nations, New York, 1958.
12. W. E. Shoupp, R. J. Coe, and W. C. Woodman, The Yankee Atomic Electric Plant, in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*, Vol. 8, pp. 492-507, United Nations, New York, 1958.
13. V. A. Elliott, R. D. Maxson, V. D. Nixon, and J. W. Merryman, The Dresden Nuclear Power Station, in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*, Vol. 8, pp. 508-534, United Nations, New York, 1958.
14. T. E. Cole, The Oak Ridge High Flux Isotope Reactor Design and Initial Operation, in Seminar on Intense Neutron Sources, USAEC Report CONF-660925-1, pp. 183-213, August 1967.
15. D. N. Shaftman and R. P. Savio, The Argonne Advanced Research Reactor (AARR), in Seminar on Intense Neutron Sources, USAEC Report CONF-660925-1, pp. 215-259, August 1967.
16. This description is a summary by the author (B. Spinrad) of remarks presented orally by R. J. Breen of Bettis, at the panel cited in Ref. 4. Any errors are the responsibility of the summarizer.
17. S. Kaplan, Synthesis Methods in Reactor Analysis, in *Advances in Nuclear Science and Technology*, Vol. 3, pp. 233-264, Academic Press Inc., New York (1966).
18. E. L. Wachspress and M. Becker, Variational Synthesis with Discontinuous Trial Functions, in Proceedings of the Conference on the Application of Computing Methods to Reactor Problems, USAEC Report ANL-7050, pp. 191-205, Argonne National Laboratory, May 17, 1965.
19. G. A. Sofer, A. H. Kazi, J. R. Tomonto, and P. Buck, Plutonium Production and Burnup Calculation Methods for Light-Water Moderated Reactors, in *Nuclear Materials Management*, Symposium Proceedings, Vienna, 1965, pp. 749-773, International Atomic Energy Agency, Vienna, 1966 (STI/PUB/110).
20. C. G. Poncelet, Effects of Fuel Burnup on Reactivity and Reactivity Coefficients in Yankee Core I, USAEC Report WCAP-6076, Westinghouse Electric Corporation, October 1965.
21. J. Jedruch and R. J. Nodvik, Experimentally Determined Burnup and Spent-Fuel Composition of Yankee Core I, USAEC Report WCAP-6071, Westinghouse Electric Corporation, July 1965.
22. R. F. Barry, LEOPARD—A Spectrum-Dependent Non-Spatial Depletion Code for the IBM-7094, USAEC Report WCAP-3269-26, Westinghouse Electric Corporation, September 1963.
23. C. G. Poncelet, LASER—A Depletion Program for Lattice Calculations Based on MUFT and THERMOS, USAEC Report WCAP-6073, Westinghouse Electric Corporation, April 1966.
24. B. J. Toppel and R. Avery, Some Physics Aspects of the Plutonium Experiment in EBWR, USAEC Report ANL-7019, Argonne National Laboratory, April 1965.
25. D. L. Delp, D. L. Fischer, J. M. Harriman, and M. J. Stedwell, FLARE—A Three-Dimensional Boiling-Water Reactor Simulator, USAEC Report GEAP-4598, General Electric Company, July 16, 1964.
26. M. F. Valerino and Z. R. Rosztoczy, Investigation of

Methods of Calculating Heavy-Isotope Content of Operating Fuel Elements, in *Nuclear Materials Management*, Symposium Proceedings, Vienna, 1965, pp. 775-796, International Atomic Energy Agency, Vienna, 1966 (STI/PUB/110).
27. Nuclear Performance of Power-Reactor Cores, USAEC Report TID-7672, American Nuclear Society, Sept. 26, 1963.
28. P. L. Hofmann, G. J. Busselman, and R. H. Holeman, Physics Statics Calculation Recipes for Plutonium Burners, USAEC Report HW-82043, Hanford Atomic Products Operation, Apr. 28, 1964.
29. D. R. Vondy, T. B. Fowler, and M. L. Tobias, Reactor Depletion Code ASSAULT (Two-Dimensional Multi-Neutron-Group, Diffusion), USAEC Report ORNL-TM-1302, Oak Ridge National Laboratory, March 1966.

Counting and Campbelling: A New Approach to Neutron-Detection Systems

By Glenn F. Popper

There has been a resurgence of activity in developing improved neutron-detection systems to broaden the range of coverage of a nuclear instrument channel, to avoid the effects of extreme direct-current (d-c) leakage currents caused by high-temperature operation, or to obtain better gamma discrimination. Most of this activity stems from the rediscovery of mathematical theorems applicable to random-event processes developed over a half century ago by Campbell.[1] Applying one of these theorems leads to the so-called "Campbelling" neutron-detection system that is based on the inherent "noise" in the neutron-detector signal.

In conventional detection systems, d-c response is directly proportional to the product of pulse rate and mean charge per pulse. In a Campbelling detection system (also called a mean-square voltage system), rms response is proportional to the square root of the product of pulse rate and mean-square charge per pulse. With appropriate electronics, conventional counting and Campbelling can be combined to monitor up to 10 decades of power with a single fission chamber.

A LOOK TO THE FUTURE

The counting–Campbelling neutron-detection approach shows particular promise when applied to liquid-metal fast breeder reactor systems, such as the Experimental Breeder Reactor No. 2 (EBR-II) and the Fast Flux Test Facility, in which large gamma fluxes caused by sodium activation are always present. The system also appears attractive from reliability and economic standpoints when applied to reactors in which instrument thimbles are at a premium, such as in the high-flux research reactors. With mean-square voltage systems, environmental limits on neutron detectors can be raised to include fluxes up to 10^{19} neutrons/(cm^2)(sec), integrated fluxes in excess of 10^{22} neutrons/cm^2, and temperatures to 1500°F (Ref. 2). These limits will certainly be realized as applications materialize.

Even though I know of no power reactors now using the combination of a counting and mean-square voltage system for reactor control, many will be using it soon.[3,4] Most of these will rely completely on in-core detectors for control. Others will require the more conventional out-of-core detectors. The number and type of detectors used for these systems could be reduced significantly through the use of a single fission chamber to monitor up to 10 decades of reactor power.

Additional development of log-N–period channels that respond to the true mean-square voltage signal is needed if this particular system is to become widely used. Adequate counting electronics and linear Campbell electronics are now available commercially and could be used whenever there is a need to improve the range or gamma discrimination of the more conventional instrument systems.

Mathematical Basis

The concepts and theorems developed by Campbell originated from early attempts to explain experimentally observed "noise" in the radiation from a radioactive substance, namely, Schweidler's fluctuations.[5] However, Schweidler's fluctuations had long been forgotten by the time these basic noise theorems were again restated by Campbell and Francis[6] in the mid-1940's and applied to linear systems, particularly to certain vacuum-tube circuits. The increasing teaching and application of random noise theory[7] and statistical analysis in the mid-1950's and early 1960's have obviously contributed to the use of noise theory to improve neutron-detection systems.

The mean theorem and mean-square theorem have come to be known as "Campbell's theorem," a term even Campbell denounced. Applicable to both linear and log systems, the basic postulate of these theorems is that noise consists of the superposed effects of events that are random in time, as are the atomic events with which quantum theory is concerned. It

must be stressed that the events themselves must be random in time for the theorems to be valid. In mathematical terms, random in time means that, if $N(T)$ is the chance that a single event happens in the interval T, then

$$N(T) \rightarrow \lambda T \qquad \text{as } T \rightarrow 0$$

Less accurately, the chance of an event happening during the infinitesimal interval dt is $\lambda\ dt$. Although λ depends on the nature of the events, it is independent of all previous events.

In mathematical form the mean and mean-square theorems are

$$\bar{y} = \lambda \int_0^\infty s(t)\ dt \tag{1}$$

and

$$\overline{y^2} - (\bar{y})^2 = \overline{(y - y)^2} = \lambda \int_0^\infty [s(t)]^2\ dt \tag{2}$$

where y = magnitude of the event at an instant t_0
$\bar{y}$ = mean value
$\overline{y^2}$ = mean-square value
λ = mean rate of event occurrence
$s(t)$ = function describing the shape of a single event

Toward Practical Neutron-Detection Systems

To see how these basic theorems are used in practical systems, we must first examine the system design equations and consider the potentially useful detectors.

SYSTEM DESIGN EQUATIONS

The signal from a radiation detector consists of a series of current pulses with a distribution of amplitudes and a Poisson time distribution in the average rate. The equations for the mean and mean-square detection systems become, from the basic noise theorems,[8]

$$E_{dc} = nq \int_0^\infty h(t)\ dt \tag{3}$$

and

$$E_{ms} = E_{rms}^2 = nq^2 \int_0^\infty [h(t)]^2\ dt \tag{4}$$

where E_{dc} = mean value of the system output, volts
n = average pulse rate, pulses/sec
$\bar{q}$ = mean charge per pulse, coulombs/pulse
$h(t)$ = function describing circuit response to a single pulse
E_{ms} = mean-square value of the system voltage output
E_{rms} = root-mean-square value of the system output, volts
$\overline{q^2}$ = mean-square value of charge per pulse (coulombs/pulse)^2

Equation 3 represents the mean output signal from a conventional d-c detection system, in which the response is directly proportional to the pulse rate and mean-pulse charge. Equation 4 represents the mean-square output signal from the so-called Campbell system, in which the measured response, E_{rms}, is proportional to the square root of the product of pulse rate and mean-square charge per pulse. [But note that Eq. 4 represents the alternating-current (a-c) signal component only, the mean value being subtracted by using a coupling capacitor or an a-c amplifier or by selecting $h(t)$ such that E_{dc} is zero.] The mathematically correct equation for the mean-square output is

$$E_{ms} = n\overline{q^2} \int_0^\infty [h(t)]^2\ dt + E_{dc}^2 \tag{5}$$

APPLICATIONS

The first known practical neutron-detection system based upon Campbell's theorem, which was extended to include the effect of amplitude difference in the events or pulses from a neutron detector, was made in 1962 (Ref. 8). This system used the a-c signal produced from the statistical fluctuations in the average pulse rate from a fission counter to generate outputs proportional to the log of the neutron flux.

DISCRIMINATION

Use of this noise signal improves the inherent discrimination against gamma pulses (which have a smaller amplitude than pulses produced by neutron interactions). Although this type of discrimination was patented in 1959, it was not applied to any known usable system.[9]

The design equations show that the d-c voltage is proportional to the mean value of charge per pulse and the mean-square voltage is proportional to the mean-square value of charge per pulse. Thus the larger neutron pulses play a greater part in determining the mean-square voltage than the mean voltage. Moreover, because of the subtraction of the d-c component from the total signal, cable leakage currents at high temperatures no longer are important in Campbelling systems.

DETECTORS

Two types of detectors can be considered for use in mean-square systems. They are the fission-counter chamber and the boron-coated ionization chamber. Each has advantages and disadvantages. With the fission counter, there are two kinds of unwanted pulses: (1) the alpha pulses produced by the natural radioactive decay of the fissile material and (2) the beta pulses produced either by detector activation or by external gamma flux. The boron-coated detector generates ionizing alpha particles from neutron interaction with the boron; the charge liberated is an order

of magnitude smaller than that from the fission detector. When this kind of detector is used, the only unwanted pulses are from beta particles caused by detector activation or by an external gamma flux, but noise at the amplifier input may become the lower limiting condition. On the other hand, proportional-counter-type detectors cannot be considered for either mean or mean-square systems. They rely on gas multiplication of the pulse, and thus their output depends critically on the polarizing supply voltage.

Fission-Counter Chambers. Figure 1 shows the calculated approximate ranges of neutron flux that can be covered with a standard fission counter operating in the presence of a gamma flux (1) as a pulse counter, (2) in a mean system, and (3) in a mean-square voltage system. The shaded areas represent the measurable ranges of neutron flux. The limit lines (solid) show where the indicated output is 10% higher than the signal due to neutrons alone.

The pulse-counting system can cover five to six decades of neutron flux in a gamma flux approaching 10^7 r/hr. The lower limit of usefulness is determined by allowable statistical fluctuations in the count rate; the upper limit is determined by the resolving time of the system and resulting counting loss.

When the same detector is used in a mean system, the lower neutron-flux limit is set either by the alpha pulse contribution or by the gamma flux; the upper limit is determined by the maximum mean current of 2 ma allowable for this type of chamber to maintain adequate chamber saturation with maximum allowable polarizing potential applied.

When the fission counter is operated in a mean-square voltage system, a much higher gamma flux can be tolerated and a much lower alpha limit is achieved as compared to a mean system.

Boron-Coated Chambers. Curves similar to those of Fig. 1 could be constructed for a boron-coated ionization chamber; the following three conclusions would be reached: (1) the detector cannot be used effectively as a pulse counter, (2) the gamma discrimination is slightly worse and the maximum range slightly extended (compared to a fission chamber) in a mean system, and (3) the discrimination becomes considerably worse and the maximum range only one decade improved as compared to a fission chamber operating in the mean-square mode.

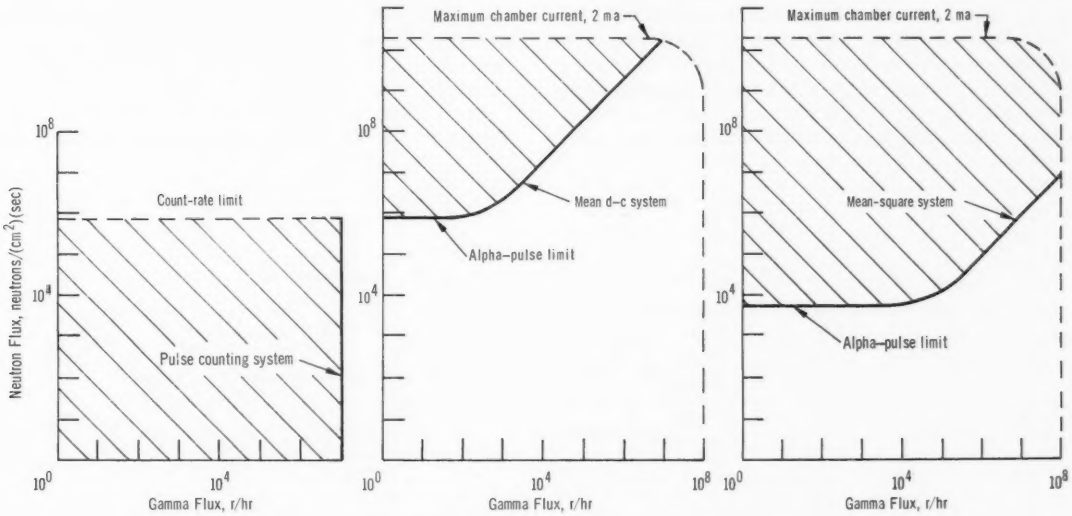


Fig. 1 Useful ranges (shaded areas) of neutron flux measurable by a fission counter operated in three modes: (1) pulse counter, (2) mean d-c system, and (3) mean-square voltage system.

System Descriptions and Components

A simplified diagram of a mean-square voltage neutron-detection system is shown in Fig. 2. It

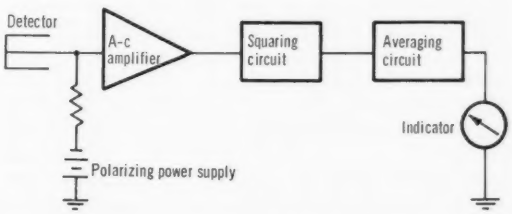


Fig. 2 Component arrangement for a mean-square voltage neutron-detection system.

consists of a detector that generates pulses, a polarizing power supply, an a-c amplifier, a squaring device, and an averaging circuit. The output of the averaging circuit is given by Eq. 4.

LOG-N CHANNELS

There are several known approaches of applying the Campbelling method to log-N channels. Figure 3 shows the first complete detection system based on Campbell's theorem.[8] The amplifier was designed to be used with a fission chamber in a system capable of covering the six decades from 10^4 to 10^{10} neutrons/$(cm^2)(sec)$. The fission chamber, a Westinghouse WL-6376, was designed for operation over a neutron-flux range of 2.5 to 10^{10} neutrons/$(cm^2)(sec)$. The system shown develops a voltage across an input circuit, the d-c component being removed by a band-pass preamplifier. The remaining a-c signal is amplified and rectified, and the circuit produces an output proportional to the logarithm of the neutron flux. Differentiation of this level output then gives a signal inversely proportional to the reactor period.

In a similar system inspired by this early work, a tuned amplifier is used (Fig. 4) in place of the band-pass amplifier.[10] This circuit uses the neutron detector and input-cable capacitance in a high-Q tuned circuit at the input followed by a linear amplifier, logarithmic amplifier, and final rectifier. The logarithmic amplifier operates on the a-c signal from the tuned circuit and drives directly a very-low-level signal rectifier; thus the need for high-accuracy amplification after the logarithmic amplifier is eliminated. Sufficient signal is developed to drive indicators and recorders; the high-level output required to drive the following differentiating amplifier for period indication can be obtained but is less stable and accurate.

Both these log-N systems, as well as a later version,[11] use rectifiers to produce the final average

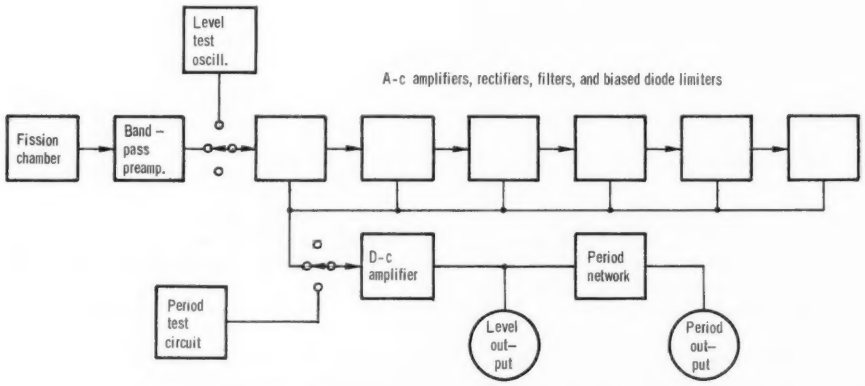


Fig. 3 The range 10^4 to 10^{10} neutrons/$(cm^2)(sec)$ was covered by the first complete detection system based on Campbell's theorem.[8]

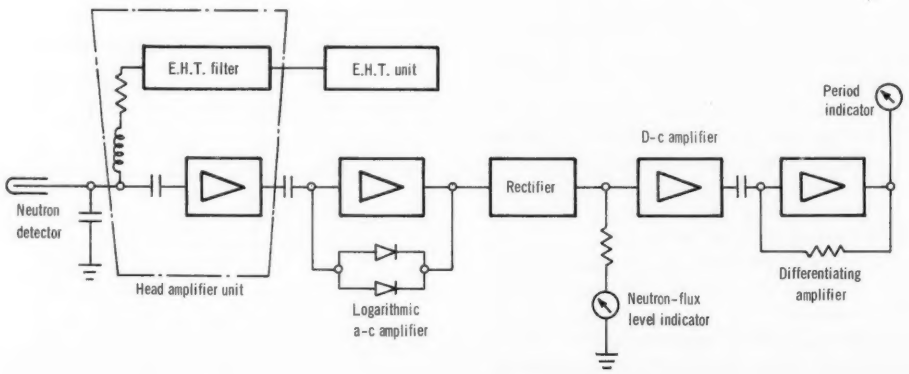


Fig. 4 An a-c logarithmic amplifier system.[10]

output, but the systems do not at any time square the fluctuating signal. This type of average-magnitude rectification is related to the true mean-square signal by a constant when the pulses are well overlapped.[8] Well-overlapped pulses are those where the product of pulse rate and pulse width exceeds unity or are above the region where the individual pulses can be resolved. Averaged-magnitude rectification is acceptable in a system where a single detector is not used to cover 10 decades of neutron flux.[4,12] However, as can be seen in Fig. 5, this type of rectification has

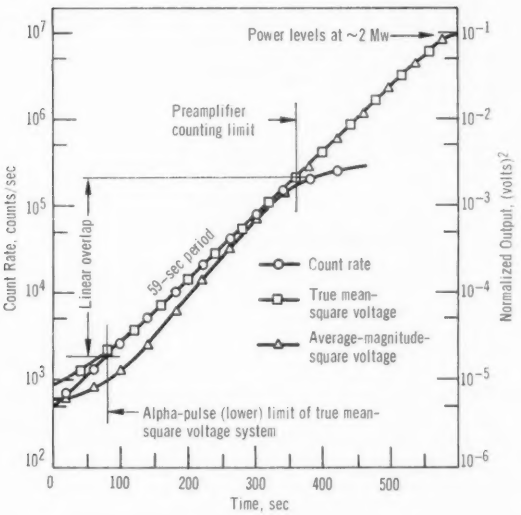


Fig. 5 Differences in response of a true mean-square system and average-magnitude-square systems are apparent in data taken while EBR-II was on a constant period.[12]

proved to be completely unacceptable when used in a wide-range system, particularly in the region of overlap between counting and Campbelling. These data, taken at EBR-II while the reactor was on a constant period, show quite clearly the differences between a true mean-square signal and an average-magnitude-square signal.

In addition to U. S.[3] and U. K.[10] efforts, the mean-square voltage approach is also beginning to be used in France[13]—in a system with an uncompensated ion chamber. The neutron-to-gamma discrimination is improved by a factor of 100 compared to a mean-current system. An experimental logarithmic amplifier has been developed for the system.

MEASURING MEAN-SQUARE VOLTAGE

How is the true mean-square voltage output from a typical system measured? Two schemes are used: (1) a circuit that actually squares the signal continuously in time and averages the squared signal and (2) the more conventional true mean-square voltmeter that uses a heating circuit as a squaring device. This voltmeter measures the power (E^2/R) of the input signal by producing heat near a thermocouple. Thermocouple output is then a measure of the mean-square voltage. Although these two types of voltage detectors give exact results at all pulse rates, the average-magnitude detection circuit will not give the correct result at all pulse rates.

True mean-square detectors are usually used in linear systems and therefore require switching to cover the full range of neutron flux. The need for log-N and period channels in conventional reactor-control systems should generate enough interest to develop the first true mean-square log-N–period intermediate-range channels.

Wide-Range Linear Systems

The full-range in-core instrumentation program at General Electric Company has probably contributed the most to mean-square voltage neutron-detection systems.[3,14–29] This program, initiated about 5 years ago,[14] was intended to develop systems and detectors to monitor power distribution in a reactor core at temperatures to 1000° F and neutron fluxes to 10^{14} neutrons/(cm^2)(sec). At first, detectors were the main concern of the program,[14–17] but by June 1964 a full-range in-core monitoring system had essentially been developed.[22]

DESIGN DESCRIPTION

This system was capable of covering the range 10^4 to 5×10^{13} neutrons/(cm^2)(sec) inside a reactor core for noncontrol applications at 550° F and in the presence of large gamma backgrounds, a significant advance in in-core neutron-detection technology. Two channels made this possible: (1) a counting channel and (2) a Campbelling (mean-square or variance-signal) channel.

Counting Channel. The counting channel consists of an in-core ion chamber integrally welded to a triaxial cable that has favorable transmission qualities. The cable terminates in the line characteristic impedance; a high-gain-current pulse amplifier produces a signal that is processed to give either linear or log count-rate indications.

Campbelling Channel. The Campbelling channel uses a separate ion chamber welded to a low-capacity cable. The detector-cable assembly couples to an a-c amplifier; output is fed to a true mean-square voltmenter. Only a linear indication is obtainable from this system; period information cannot be generated directly.

The gamma limitation was 6×10^6 r/hr for the counting channel, although larger gamma fluxes can be tolerated if the resulting counting losses can be accepted. For the mean-square voltage channel, the

neutron-to-gamma signal ratio was 0.4 r/hr/[neutrons/(cm^2)(sec)]. This compares to a ratio of 2.2×10^{-3} for the same chamber operating in the d-c mode.

In-Core Ion Chambers. Until 1964, in-core ion chambers were limited to power-range measurements [10^{11} to 5×10^{13} neutrons/(cm^2)(sec)], where the chamber was operated in strictly the mean-current or conventional d-c mode. Fission chambers were not used in the startup range as counters because it was not known how pulses could be transmitted from the chamber to an out-of-core amplifier. Application of transmission-line theory solved this problem. Moreover, ion chambers had not been used in the intermediate range because count rates were too large to separate pulses and because leakage and gamma currents swamped out the neutron-produced currents. Application of Campbell's theorem and measurement of the mean-square voltage, rather than of the d-c signal, have solved the problem.

AVAILABLE HARDWARE

A further extension of the Campbelling channel effort led to the development of wide-range linear-system hardware that now is commercially available.[3,23–29] An excellent summary of this work is presented in Ref. 26, which describes the analytical predictions and experimental results of the nuclear instrumentation program. The program, which is concerned with the development of full-range reactor control that incorporates either in-core or out-of-core ion chambers, has produced a system composed of two electronic subsystems. Counting techniques were used for source and lower intermediate-range neutron fluxes; Campbell or mean-square voltage techniques were used for intermediate- and power-range fluxes. Furthermore, the program investigated the problems associated with (1) an in-core detector for counting systems, (2) an in-core detector for Campbelling systems, and (3) transmission lines. Also included were problems unique to a two-channel full-range instrumentation system. A system compatible with either in-core (Fig. 6) or out-of-core (Fig. 7) detectors has been realized, and hardware has been assured for a broad range of applications. Ranges for both arrangements are shown in Fig. 8.

Single-Detector Systems

The possibility of using a single neutron detector to monitor up to 10 decades of reactor power has been suggested.[26] In the first such system (Fig. 9), a standard fission counter operating at 300 volts provided the signal to a preamplifier.[12] Preamplifier output was connected directly both to a linear pulse amplifier–discriminator and count-rate meter and to a true rms voltmeter. The mean-current signal was also measured with a standard picoammeter.

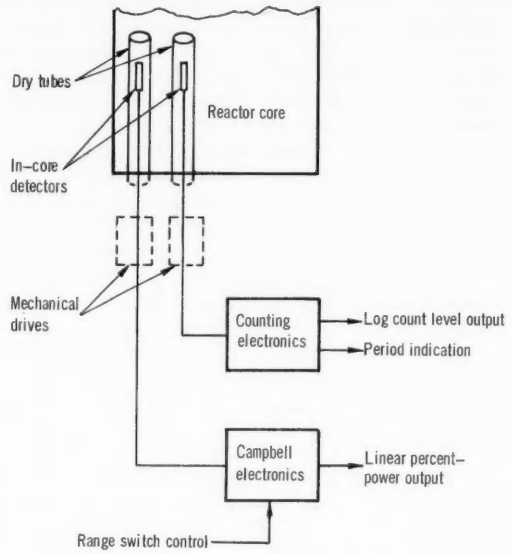


Fig. 6 In-core log count-rate monitors and multirange mean-square-voltage monitor systems.[26]

During the experiment an average-magnitude voltmeter was also used to measure preamplifier output.

OUTPUT SIGNALS

A conventional counting technique covered the source and low-power ranges, but a Campbelling system was used to monitor the intermediate- and high-power ranges. Sufficient linear overlap of the two output signals has been verified experimentally to ensure continuous neutron-flux indications. A typical response of this system to neutron flux is presented in Fig. 5, and another is shown in Fig. 10. No attempt was made to develop the sophisticated electronics that are required for a reactor prototype con-

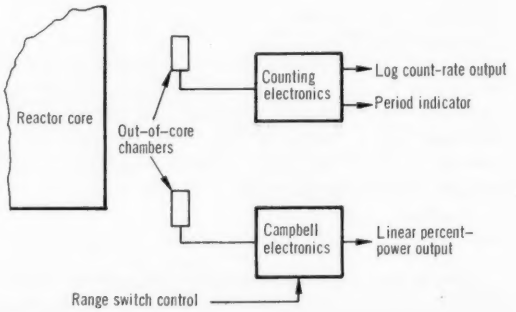


Fig. 7 Out-of-core log count-rate monitors and multirange mean-square-voltage monitor systems.[26]

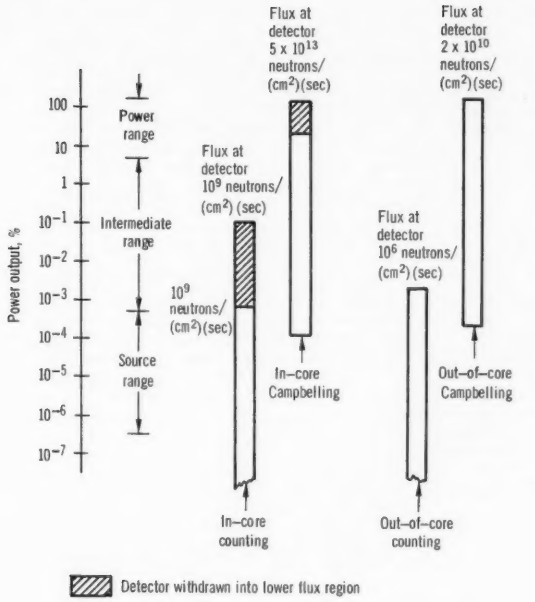


Fig. 8 Ten decades of power are covered by in-core and out-of-core counting and Campbelling systems shown in Figs. 6 and 7 (Ref. 26).

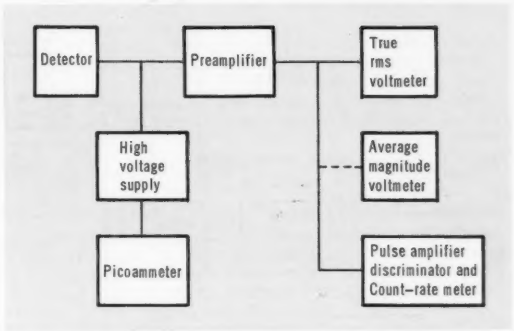


Fig. 9 Single-detector system covers the source and low-power ranges with conventional counting techniques and the intermediate- and high-power ranges with a Campbelling system.[12]

trol system. However, a complete description of the work on this single-detector system is given in Ref. 4.

The need for log-N–period channels has been almost too firmly established in conventional reactor-control systems to ignore. Trenholme and Keefe,[11] in a step toward such channels, have assembled a system (Fig. 11) that is similar to that described in Ref. 8, except that a counting channel has been added and a single detector utilized. An average-magnitude rectifier circuit is used to obtain the mean-square voltage output signal; the results reconfirm that there is essentially no overlap between the counting and log-N–period channels.[4,12]

Practical Extensions

Because plant electrical noise sources can add a component to the mean-square voltage appearing at the amplifier output, pickup must be carefully controlled—usually by the normal practices of shielding, grounding, and isolation. When these normal techniques do not adequately reduce plant noise, a possible solution is described in Ref. 30 by De Lorenzo, who discusses the excellent noise-rejection characteristics when a fission detector is connected to a pulse transformer driving a balanced transmission cable. When combined with a tuned circuit input for Campbelling, this type of drive could improve the signal-to-noise ratio. A single detector could still be used for both counting and Campbelling. This approach appears to be a practical extension of present concepts.

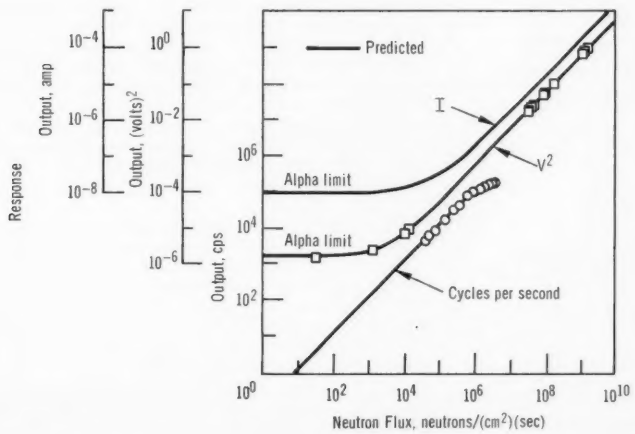


Fig. 10 Response of a single detector operated as a pulse counter and also in a Campbelling system.[4]

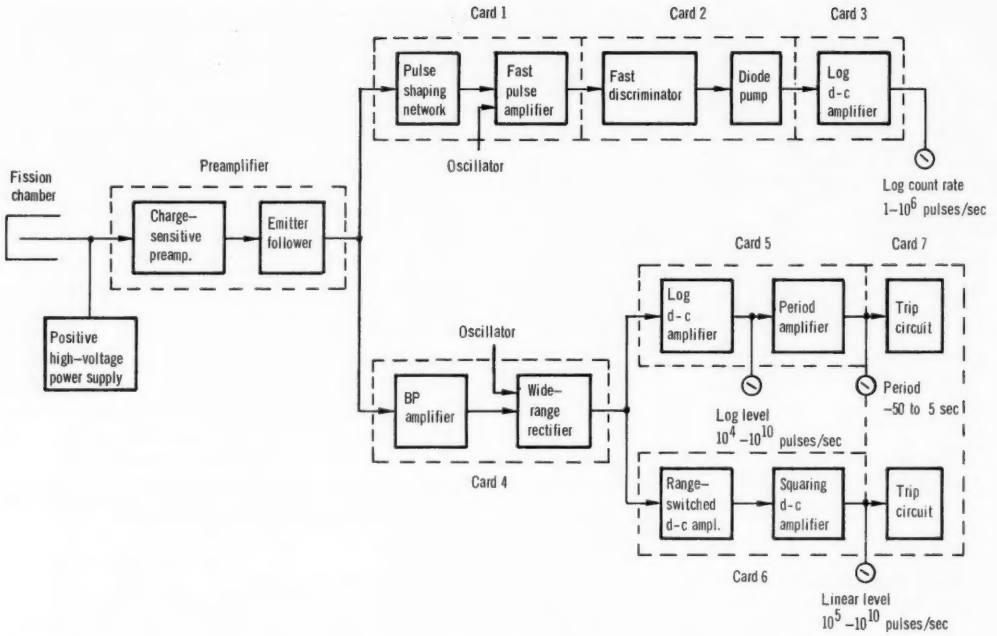


Fig. 11 Log-N–period channels using single detectors and counting and Campbelling systems.[11]

If further improvement in gamma discrimination is required, the neutron flux can be determined by measuring moments higher than the second (Campbelling). The equations for higher moments have been investigated comprehensively,[31] and it appears that a third-moment system is the highest practical one. Predictions are for an improvement of 10^3 in gamma discrimination over a Campbell system and of 10^6 over a d-c system.

Finally, in many reactor applications the lower limit of neutron detection for a mean-square voltage channel will be set by the alpha pulse component from the fissionable material in the chamber itself. Techniques such as internal alpha shielding of the fissile material and isotopic-material selection may reduce this unwanted signal. Then a wider range of neutron-flux monitoring and greater signal overlap could be achieved.

References

1. N. R. Campbell, The Study of Discontinuous Phenomena, *Proc. Cambridge Phil Soc.*, 15: 117, 310, 513 (1909–1910).
2. R. A. DuBridge, Campbell Theorem—System Concepts and Results, *IEEE (Inst. Elec. Electron Eng.), Trans. Nucl. Sci.*, NS-14(1): 241-246 (February 1967).
3. J. J. Forster, In-Core Neutron Flux Instrumentation, *IEEE (Inst. Elec. Electron. Eng.), Trans. Nucl. Sci.*, NS-12(6): 8-14 (December 1965).
4. G. F. Popper, W. C. Lipinski, and J. M. Harrer, A Wide Range Gamma Compensated Neutron Flux Detection System, USAEC Report ANL-7224, Argonne National Laboratory (to be published).
5. E. V. Schweidler, Premiére Congrés International Pour l'Étude de la Radiologie at de l'Ionisation, Liège, France, 1905.
6. N. R. Campbell and V. J. Francis, A Theory of Valve and Circuit Noise, *J. Inst. Elec. Eng. (London)*, 93, Pt. III (21): 45–52 (January 1946).
7. J. S. Bendat, *Principles and Applications of Random Noise Theory*, John Wiley & Sons, Inc., New York, 1958.
8. D. A. Gwinn and W. M. Trenholme, A Log-N and Period Amplifier Utilizing Statistical Fluctuation Signals from a Neutron Detector, *IEEE (Inst. Elec. Electron. Eng.), Trans. Nucl. Sci.*, NS-10(2): 1-9 (April 1963).
9. R. M. Lichtenstein, Radiation Measuring Instrument, U. S. Patent No. 2,903,591, Sept. 8, 1959.
10. E. P. Fowler and R. W. Levell, The Use of the Current Fluctuations from a Neutron Detector with Logarithmic Amplifiers as a Measure of Neutron Flux Levels, British Report AEEW-R-375, June 1964.
11. W. M. Trenholme and D. J. Keefe, A Neutron Flux Measuring Channel Covering Ten Decades of Reactor Power with a Single Fixed Position Detector, *IEEE (Inst. Elec. Electron. Eng.), Trans. Nucl. Sci.*, NS-14(1): 253-260 (February 1967).
12. G. F. Popper, W. C. Lipinski, and J. M. Harrer, Ten Decades of Continuous Neutron Flux Monitoring from a Single Fixed Position Detector, *Trans. Amer. Nucl. Soc.*, 9(1): 316-317 (June 1966).
13. Y. Plaige and R. Quenee, The Use of Campbell's Method in Reactor Nuclear Control Instrumentation, *IEEE (Inst. Elec. Electron. Eng.), Trans. Nucl. Sci.*, NS-14(1): 247-252 (February 1967).
14. R. A. DuBridge, In-Core Power Monitoring of Nuclear Reactors, USAEC Report GEAP-3914, General Electric Company, April 1962.

15. A. C. Duckart, Telemetering. In-Core Instrumentation Development, First Topical Report, Task II, Phase I, USAEC Report GEAP-4026, General Electric Company, Atomic Power Equipment Department, March 1962.
16. R. A. DuBridge, Detectors for In-Core Power Monitoring. In-Core Instrumentation Development Program, Quarterly Progress Report, January–March 1963, USAEC Report GEAP-4222, General Electric Company, Atomic Power Equipment Department, April 1963.
17. R. A. DuBridge, Detectors for In-Core Power Monitoring. In-Core Instrumentation Development Program, USAEC Report GEAP-4304, General Electric Company, Atomic Power Equipment Department, June 1963.
18. R. A. DuBridge, The Feasibility of Using In-Core Ion Chambers To Cover the Complete Reactor Neutron Flux Range from Startup to Full Power. In-Core Instrumentation Development Program, Quarterly Progress Report, June–September 1963, USAEC Report GEAP-4386, General Electric Company, Atomic Power Equipment Department, October 1963.
19. R. A. DuBridge, The Feasibility of Using In-Core Ion Chambers To Cover the Complete Reactor Neutron Flux Range from Startup to Full Power. In-Core Instrumentation Development Program, Quarterly Progress Report, September–December 1963, USAEC Report GEAP-4483, General Electric Company, Atomic Power Equipment Department, January 1964.
20. R. A. DuBridge, Mean Square Voltage Fluctuation Measurements with Neutron Sensitive Ion Chambers, *Trans. Amer. Nucl. Soc.*, 6(2): 306-307 (November 1963).
21. R. A. DuBridge, Application of Counting Techniques to In-Core Ion Chambers. In-Core Instrumentation Development Program, Quarterly Progress Report, January–March 1964, USAEC Report GEAP-4604, General Electric Company, Atomic Power Equipment Department, April 1964.
22. R. A. DuBridge and W. K. Green, A Full-Range In-Reactor-Core Neutron Monitoring System Employing Ion Chambers, USAEC Report GEAP-4643, General Electric, Company, Atomic Power Equipment Department, June 1964.
23. J. P. Neissel, Theory of the Campbell System of Reactor Instrumentation, In-Core Instrumentation Development Program, Quarterly Progress Report, July–September 1964, USAEC Report GEAP-4747, General Electric Company, Atomic Power Equipment Department, October 1964.
24. J. P. Neissel and W. K. Green, General Electric Company, Atomic Power Equipment Department, March 1965.
25. L. R. Boyd et al., Reactor Control Systems Based on Counting and Campbelling Techniques. Wide-Range Instrumentation Development Program, Seventh Quarterly Progress Report, February–April 1965, USAEC Report GEAP-4862, General Electric Company, Atomic Power Equipment Department, May 1965.
26. R. A. DuBridge et al., Reactor Control Systems Based on Counting and Campbelling Techniques. Full Range Instrumentation Development Program, Final Progress Report, USAEC Report GEAP-4900, General Electric Company, Atomic Power Equipment Department, July 1965.
27. J. P. Neissel, Design Equations for a Neutron Flux Monitor Using Current Fluctuations from a Detector, *Trans. Amer. Nucl. Soc.*, 8(1): 105 (June 1965).
28. R. A. DuBridge, Application of In-Core Detectors to the Startup of Large Boiling Water Reactors, *Trans. Amer. Nucl. Soc.*, 8(1): 105 (June 1965).
29. R. A. DuBridge, J. P. Neissel, and L. R. Boyd, Application of In-Core Detectors to the Startup of Large Boiling Water Reactors, *IEEE (Inst. Elec. Electron. Eng.), Trans. Nucl. Sci.*, NS-13(1): 438-442 (February 1966).
30. J. T. De Lorenzo, A Neutron Counting System with a Fission Detector in a Flexible, Balanced Line, *IEEE (Inst. Elec. Electron. Eng.), Trans. Nucl. Sci.*, NS-14(1): 261-270 (February 1967).
31. I. Kliger, Argonne National Laboratory, personal communication, January 1967.

Vibration of Reactor Core Components

By Martin W. Wambsganss, Jr.

High-velocity coolant flowing through a reactor core is a source of energy that can induce and sustain vibration in reactor core components. In fact, there have been numerous flow-induced vibration problems, notably in the EBR-II, GETR, Big Rock Point, Yankee, SPERT-III, and MSRE reactors.[1–13] These problems included: (1) excessive oscillations, which resulted in excessive wear, fretting, and fatigue; (2) incorrect flow distribution; (3) broken specimen holders; (4) loss of thermowells; and (5) excessive vibration in heat exchangers.

In the past, trial-and-error corrections have successfully solved many vibration problems associated with reactor design, sometimes without knowing the precise mechanisms causing the vibration.

In the future, as both power densities and coolant velocities increase, core internals will be subjected to more severe conditions. Further, the more sophisticated and optimal design may not allow for the trial-and-error changes required to reduce or eliminate the objectionable vibrations. Thus the philosophy of "remedy later" is becoming less and less practicable; complex vibration problems must now be solved during the concept and design stages.

Many structural problems are unique to the nuclear reactor field. This is due, for example, to the small clearances between components, and the adverse operating environment consisting of high temperatures, thermal gradients, and high-velocity coolant flows, in addition to the radiation field. Therefore the existing technology must be extended to satisfactorily explain and predict even presently observed vibrational phenomena associated with reactor core components.

Origins of Vibrational Problems

With the exception of the control rods, the in-core components of a reactor are, in general, stationary elements with no designed motion functions. During normal reactor operation the control rods themselves might be thought of as quasi-stationary elements. The reactor vessel of a land-based reactor is not subject to any motion if we neglect earthquakes and similar environmental disturbances. Since vibration problems are generally associated with motion, a cursory review may lead to the conclusion that there are no vibration problems at all.

But, because every structural component possesses mass and elasticity, every component has some potential for mechanical vibration. To realize this potential requires that there also be a compatible source of energy and coupling. For reactor core components the primary energy source, as well as the coupling means, is the coolant flowing through the core. An example of an additional energy source and coupling path is that of pump oscillations and structure–structure interaction. However, even in this example the coolant remains the essential feature because it transmits the pump oscillations and maintains the vibrations.

Recent Vibrational Problems

A survey of the recent occurrences of vibrational problems shows that the majority are flow-induced. The collapse of reactor fuel plates subjected to water flow was observed as far back as 1948 (Ref. 14). More recent occurrences of flow-induced vibration problems have been reported in connection with the following reactors: EBR-II,[1] GETR,[2,3] Big Rock Point,[4–7] Yankee,[8–10] SPERT-III,[3,11] and MSRE.[12,13]

EBR-II

An analysis of the EBR-II at various power levels[1] revealed the existence of a self-excited 10-cps power oscillation over the range 20 to 45 Mw(t). The peak amplitude of the oscillation was 112 kw at 45 Mw. Experiments eliminated various potential causes and led to the following proposed mechanism, due to the flow-induced vibration of fuel subassemblies. At low power (< 20 Mw), small but definite clearances exist between the fuel subassemblies, and for analytical purposes the core region is mechanically uncoupled. The rapid flow of coolant through the individual subassemblies induces vibrations that are manifested in the power output as Gaussian noise. However, power increases

cause the subassemblies to bow; these bowing effects reduce the clearances first at the top and then, as the bowing continues toward the center, cause wall-to-wall contact. Under these conditions the flow-induced vibrations become increasingly coherent as the power, and hence mechanical coupling, increases.

The calculated natural frequency of a standard EBR-II subassembly in sodium is 5.2 cps. Since one complete vibration of a coupled subassembly system produces two reactivity perturbation cycles, the 10-cps power oscillation may be explained in terms of a simple 5.0-cps coupled vibration of the core.

GETR

Hydraulically induced core vibrations in the GETR caused core damage and period oscillations.[2,3] Damage was defined as an excessive wear rate of core-component spacer pads which provided mutual lateral support for the upper ends of the components.

Motion pictures taken of the core under full-flow conditions showed that hydraulic forces acting on the control-rod guide-tube channels caused them to vibrate with a frequency of approximately 8 cps at an amplitude of $\frac{1}{2}$ in. The reactor log-N-period instrumentation was sensitive to these vibrations, and, as the component spacer pads eroded, the recorded log-N-period oscillations increased in amplitude. Two design changes made to eliminate this vibration problem were: (1) bolting the free ends of the guide tubes to the primary coolant-flow distributor and (2) changing the spacer-pad material from aluminum to stainless steel.

BIG ROCK POINT

Excessive vibration due to an inadequate flow distribution of the coolant as it entered the reactor vessel[4,7] was the cause of the loosening and dislodging of the bolts that connected the fuel channels to the channel support tubes in the Big Rock Point Nuclear Power Plant. Accelerometers and strain gauges were attached to channels in the reconstituted reactor core, and readings were obtained for various conditions of coolant flow. These values pinpointed the coolant-inlet-flow distribution as the cause of the vibration problem. A baffle was designed and installed to give a more equal distribution of the coolant flow entering the vessel. Additional modifications of the 72 fuel channels, to prevent loosening of the bolts and separation of the channels and support-tube assembly, were also made.

Operators of the Big Rock Point reactor power plant observed neutron-flux oscillations that were attributed to a mechanical vibration.[5-7] During shutdown following a reactor scram, an inspection revealed that 6 of the 12 studs holding the thermal shield had failed. The progressive failure of these hold-down studs produced a rotational mode of vibration of the thermal shield about the horizontal axis. Neutron-flux oscillations at the frequencies observed could be directly related to this mode of thermal-shield motion. Demonstrations showed that the presence of a flowing energy source caused the thermal shield to "break into" sustained self-excited vibrations. The flow-induced vibration of the thermal shield was a result of the early flow-induced vibration in support-tube-and-channel assemblies which caused fretting between the alignment pins and the mating slot. Fretting produced clearances which permitted movement of the thermal shield and led to the eventual fatigue failure of the studs.

YANKEE

During the refueling of cores 1 and 2 of the Yankee reactor,[8,10] evidence of hydraulic flutter of control-rod blades and stationary shim rods was observed. Also discovered at the same time was the failure of specimen holders due to buffeting from hydraulic flow. This was a problem of more serious consequence since some of the specimen holders broke and lodged in places in the reactor vessel where they were difficult to locate and remove.

During the Yankee core 4 cycle,[8-10] prompted by vibration problems in other reactors, the Yankee Atomic Electric Company asked for an engineering review of the potential vibration problems of the Yankee reactor internals. The review indicated that no vibration problem should exist. For other reasons, the 25-ft-long, 9-ft-OD barrel with 0.040-in.-radial clearance was pulled out for inspection. The relatively small clearance of the large-size barrel resulted in very little hydraulic-flow-induced barrel vibration, and examination showed no cyclic overstressing of the supporting gussets or flange weld.

Additional incidences of flow-induced vibration problems were reported, but they were not directly associated with the reactor internal components.

SPERT-III

The SPERT-III reactor lost a thermowell in the primary piping system as a result of flow-induced vibration.[3,11] Earlier fatigue failures of in-core instrumentation were also attributed to vibration of small parts exposed to the coolant flow.[3] Strain-gauge tests showed that the 9- and 6-in. wells were excited at their natural frequencies, whereas 5-in. wells showed essentially no excitation.

MSRE

Difficulties with excessive vibration in heat exchangers at several plants (Fermi and Hallam) prompted a review and subsequent testing of the heat-exchanger design for the MSRE.[12] In this plant, heat from the fuel-bearing molten salt is transferred to a coolant salt in the heat exchanger. At flow rates above 900 gal/min, noises were heard coming from the MSRE heat exchanger. Later tests[13] showed that the heat-exchanger

tubes vibrated excessively at flow rates above 800 gal/min. Tube vibration was eliminated by attaching tight-fitting spacer bars to the baffle plates between the rows of tubes.

Occurrences such as these have prompted many recent vibration studies related to specific reactor designs and have aroused the concern of review committees. Although in many cases the causes of the vibrations have been identified and the vibration problems have been solved by redesign, various mechanisms involved, for example, that of energy extraction from the flowing coolant, are not completely understood.

Characteristics of Vibrations

Although vibrations can be useful, or merely a nuisance, the destructive vibrations that can affect rods, plates, and panels present the chief problems in structural analysis for reactor designers. The analyst must consider the potential of each component to be forced or excited by the flowing coolant (whether liquid or gas) and must predict the dynamic behavior (stability and/or response) of each component.

FORCED AND SELF-EXCITED VIBRATIONS

Sustained vibrations can generally be classified as either forced or self-excited. In a forced vibration the forces exist independent of structural motion. However, a self-excited vibration is one in which the forces are a function of the displacement or velocity of a structure and exist only when the structure is not in its equilibrium position. Self-excited vibrations can be initiated by various transient disturbances or thermal effects.

LINEAR AND NONLINEAR VIBRATIONS

Actually, all vibrations are nonlinear. However, if the displacements are small, a linear analysis will usually give a satisfactory solution. A nonlinear analysis is required if large displacements can be expected or if an attempt is made to describe phenomena inherent only in nonlinear systems (for example, the limit cycle associated with self-excitation).

STABILITY AND RESPONSE ANALYSES

Vibration analysis may be divided into the consideration of stability and response problems. In a stability analysis the problem is to determine whether conditions exist that cause structural response to diverge and, if so, to find these conditions (stability limits) in terms of the system parameters. In response analysis the problem is to predict the actual motion of the structure.

The stability analysis is generally simple and often supplies sufficient information to consider the vibration problem solved. If the actual motion of the structure is desired, or if we wish to know what happens to the structure under unstable conditions, the response problem must be solved. This requires knowledge of the damping mechanism and associated damping laws (neither of which is accurately known or predictable) and often necessitates solving a nonlinear differential equation.

FLUID–ELASTIC EFFECTS

Mechanical vibrations are classically analyzed as if occurring in a vacuum; this is most often a reasonable assumption when the fluid environment in which the system vibrates is a gas. However, when the fluid medium is a liquid, which is usually the case within a reactor, the effect of the fluid becomes significant and what might be termed a "fluid–elastic" analysis is required. The fluid–elastic analysis necessitates drawing from the field of fluid dynamics as well as from fields related to the theory of elasticity and mechanical vibrations. The additional discipline of stochastic processes may be required to account for the randomness characteristic of most fluid–dynamic phenomena, for example, turbulence.

Knowledge of the stability and response of flow-induced vibrations has progressed to the point that the more significant contributions related to reactor core components can be enumerated and discussed in some detail. These contributions can be classified under the following headings: (1) vibration of rods in cross flow, (2) vibration of rods in parallel flow, and (3) flat-plate vibration.

Vibration of Rods in Cross Flow

A primary cause of transverse vibrations perpendicular to the flow velocity is vortex shedding. The frequency at which vortices are shed from a cylinder is represented in nondimensional form by the Strouhal number, or reduced frequency, defined as

$$k = \frac{\omega d}{U} \qquad (1)$$

where ω is the vortex shedding frequency, d is the diameter of the cylinder, and U is the cross-flow velocity.

The Reynolds number, based on the diameter of the cylinder, also effects the shedding of vortices. For a cylinder, Fig. 1 gives the Strouhal number as a function of Reynolds number.[15] It can be seen that for a range of Reynolds numbers ($10^3 < \mathrm{Re} < 10^5$) the Strouhal number is approximately equal to a constant value of 1.2.

The Strouhal number for the cylinder can be defined as

$$k_{Str} = \frac{\omega_0 d}{U} \qquad (2)$$

where ω_0 is the natural frequency of the structure.

For an anticipated flow velocity, Reynolds number is calculated. The corresponding Strouhal number for that flow velocity can then be read from Fig. 1; call it k_{cr}. If k_{St} is sufficiently greater than k_{cr}, the natural frequency of vibration of the structure can be concluded to be sufficiently above the vortex shedding frequency to preclude possible resonance effects caused by coincident frequencies.

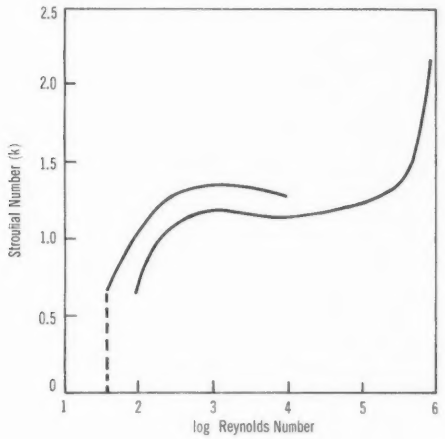


Fig. 1 For Reynolds numbers between 10^3 and 10^5, the Strouhal number k *is ~1.2, as shown by these data for cylinders.*[15]

Rather detailed reviews relating to vortex formation and excitation have been made.[16,17] Related studies, associated with heat exchangers, have been conducted by a number of investigators.[18–20]

It is difficult to predict the amplitude of vortex excitations. However, the maximum intensity of the resultant alternating force can be expressed[21] in the usual form for aerodynamic forces as

$$F = C_K \tfrac{1}{2} \rho V^2 A \sin \omega t \qquad (3)$$

The force coefficient C_K may be taken equal to 1, which implies the magnitude of the force, per unit sidewise projected area (A), is equal to the stagnation pressure of the flow ($\frac{1}{2} \rho V^2$).

Vibration of Rods in Parallel Flow

Although the vibration of rods or cylinders induced by cross flow was recognized early and has consequently received considerable attention, the study of the vibration of rods in parallel flow is less than a decade old. It should be noted, however, that the closely related problem regarding the vibration of a pipe containing a flowing fluid has been treated earlier in the literature.[22–25]

FIRST STUDIES

In 1958, Burgreen, Byrnes, and Beneforado, reported on an experimental program that demonstrated the phenomena of rod vibration induced by parallel flow.[26] They were led to the program as a result of observations made while studying the heat transfer of water flowing across a bundle of rods simulating a reactor core. This work was perhaps the earliest study motivated by a concern that parallel-flow-induced oscillations of cylindrical fuel elements may cause a mechanical design problem in a reactor.

Their studies were performed in a flow loop and involved a single rod and bundles of 13 and 37 rods on triangular lattices with various equivalent hydraulic diameters. The experiments demonstrated that oscillations occur in parallel flow at all velocities. When the rods were resonated in water rather than air, they vibrated at a lower frequency. The fact that the vibration frequencies remained relatively constant over the range of flow velocities led to the conclusion that the vibrations were self-excited rather than forced, for example, by Von Karman vortices. The general trend was an increase in amplitude of the oscillation with flow velocity. In conjunction with this experimental work, an empirical expression was developed for the displacement amplitude. This was accomplished by a dimensional analysis using a differential equation to describe the motion.

Burgreen's work was followed by independent experimental studies, some related to specific reactor designs and some proposing empirical vibration amplitude relations, in the United States,[27–31] Sweden,[32–34] France,[35] and Canada.[36,37] Quinn[28] conducted tests on single rods and multirod assemblies in both single- and two-phase flow. He developed a mathematical model that gave a solution in terms of several known nondimensional parameters. He evaluated his model by comparing trends in experimental data with trends indicated by the solution to the mathematical model. On the basis of this method of evaluation, the comparison indicated the model was in agreement with experimental trends above an undefined critical velocity. However, his model is difficult to apply.

A REFINED MODEL

The results of the experiments conducted by the Swedish and French investigators involved single rods and rod bundles in axial flow. The rod specimens were tested in a variety of geometrical arrangements and under various fluid flow conditions. Paidoussis[36] collected these experimental results and added to them Quinn's data. When used to correlate the data, the empirical expression derived by Burgreen et al. showed discrepancies up to 10 times the values attained from water tests and up to 100 times the values for superheated-steam tests.[36]

Using a force-balancing technique, Paidoussis derived a new empirical expression for the amplitude

of transverse vibration of cylindrical beams and clusters of cylinders in axial flow. He considered an element of the cylinder to be acted upon by the following forces: (1) flexural restoring force, (2) force due to longitudinal tension, (3) an inertial force, and (4) lateral fluid forces (reaction on cylinder of the force required to accelerate the fluid around it and frictional forces). Several nondimensional parameters were obtained in the resulting equation. Those variables involving the cross flow and swirl velocities, the tension, and surface roughness were neglected due to the absence of experimental data.

Plots of the reduced amplitudes against various parameters, with the remaining parameters fixed, gave the exponents of the pertinent dimensionless parameters. For example, the amplitude was approximately proportional to U^3 (U, axial flow velocity), D_h (hydraulic diameter), and $(L/D)^2$ (L/D, length-to-diameter ratio); this suggested the grouping

$$(u^2 N_R \epsilon^2)$$

where $u = (M/EI)^{1/2} UL$
N_R = Reynolds number = $\rho U D_h/\mu$
$\epsilon = L/D$
M = mass of fluid per unit length
EI = rigidity of rod

The dependence of the amplitude of vibration (δ) on $(u^2 N \epsilon^2)$ for increasing flow velocities with all other parameters constant showed the group to vary according to a power between 2/3 and 1. The average value of 4/5 was chosen as the exponent, and a factor $(1 + 2u^2)^{-1}$ was introduced to correct for discrepancies at high flow velocities. Paidoussis' resulting empirical formulation can be written

$$\frac{\delta}{D} = \alpha^{-4}\left[\frac{(u^2 N_R \epsilon^2)^{4/5}}{1 + 2u^2}\right]\left[\frac{\beta^{2/3}}{1 + 4\beta}\right] 10^{-5} \qquad (4)$$

where α is the eigenvalue of the fundamental normal mode of rod vibration, β is $M/(M + m)$, and m is mass per unit length of cylinder. Although a reasonable correlation (better than that obtained with previously postulated expressions) has been obtained, Paidoussis has pointed out: ". . . there remains a wide margin of possible error."

Spacer Effects. Later experimenters[28,31] have attempted to determine the flow-induced vibration of an assembly of fuel rods with intermediate spacers. The flow test bundles were 6 ft long with 16 rods arranged in a 4- by 4-square-pitch array. The rods were loaded with lead pellets and the ends secured to approximate fixed-end conditions. Tests were run with one, two, and four spacers and water temperatures from 70 to 150°F.

Pavlica and Marshall[29] compared their results with predictions based on Burgreen's and Paidoussis' empirical formulations. Paidoussis' correlation can be seen in Fig. 2 to agree fairly well with experimental results for 70°F water. Burgreen's correlation, however, for the same case predicted vibration amplitudes more than 250% higher than those of Paidoussis. Both correlations satisfactorily describe the change in amplitude with increasing flow velocity as being approximately proportional to the flow velocity to the 2.3 power. Experiment also showed that the vibration of a fuel assembly was similar to individual rod vibration—both vibrated near their natural frequencies.

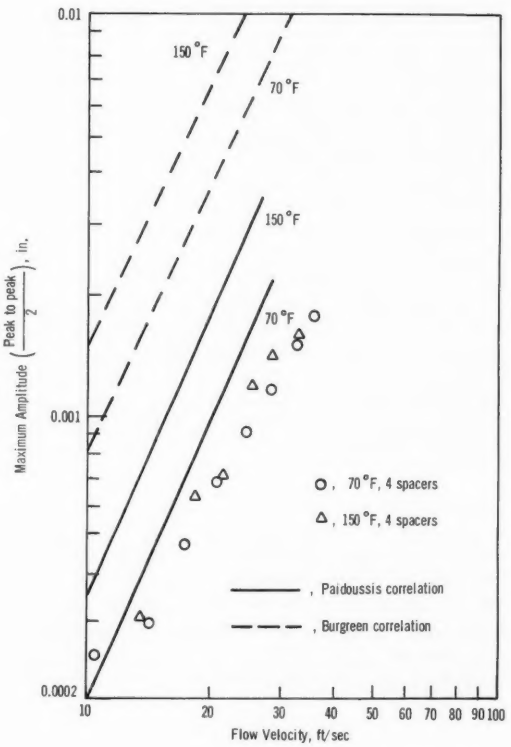


Fig. 2 Vibration amplitude increases as temperature increases.[29]

Viscosity Effects. A major discrepancy resulted when the empirical formulations of Burgreen and Paidoussis were used to correlate vibration amplitudes obtained in water tests at 150°F. Both formulations predicted a 90% increase in vibration amplitude due to the decrease in viscosity attributable to the increase in temperature.

A conclusion of Pavlica and Marshall is that Paidoussis' correlation fits the data fairly well except for the viscosity effect. A further conclu- except for the viscosity effect. A further conclusion is that additional work is required using different-

size bundles and wider temperature ranges to confirm the applicability of Paidoussis' correlation to composite fuel-bundle vibration.

Velocity Effects. More recently, Paidoussis[37,38] has extended his study of the vibration of flexible cylinders induced by axial flow. Using the differential equation of motion derived from a force balance in the lateral direction, he first solves the simpler stability problem. His approach is to assume motions of the cylinder of the form $Y(\xi)e^{i\omega t}$, where ω is a dimensionless frequency. With this form for the response, the system will be stable if the imaginary component of ω, $I(\omega)$, is positive and unstable if $I(\omega) < 0$.

The complex frequency for finite values of the flow velocity was determined by analytical and numerical methods.[38] Stability in the three lowest modes was considered for a particular case. From Fig. 3 it can be seen that small flow velocities damp free oscillations of the cylinder in all three modes $[I(\omega) > 0]$. For sufficiently high values of dimensionless flow velocity, the system becomes unstable $[I(\omega) < 0]$ in all three modes. The first-mode instability is of the buckling type $[R(\omega) = 0]$, and second- and third-mode instabilities are oscillatory $[R(\omega) \neq 0]$.

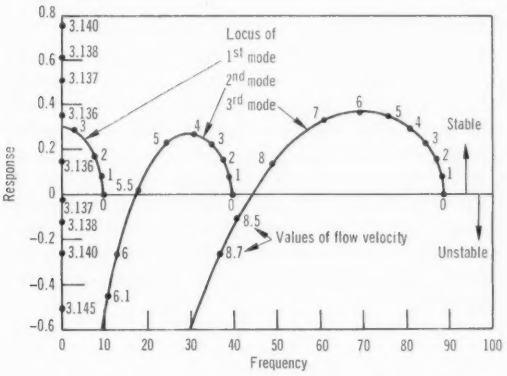


Fig. 3 Dimensionless complex frequency of the three lowest modes of a pinned–pinned cylinder as a function of dimensionless flow velocity. Points on the ordinate are part of the first-mode locus.[38]

Paidoussis verified these instabilities experimentally by using cast rubber cylinders in a flow loop.[37] However, since hydroelastic instabilities occurred at flow velocities not likely to be used in reactor coolant channels, he concluded that they need not concern reactor designers. For example, the critical flow velocities for a hollow steel cylinder of ½-in. outside diameter, 1/64-in. wall thickness, and 24-in. length in water flow are 367 and 729 ft/sec for first- and second-mode instabilities. A compressive load of 290 lb reduces these critical velocities to 160 and 649 ft/sec, which still are very high flow rates.

Paidoussis terms the response problem "subcritical vibration." The response of the cylinder was observed at all flow velocities as small-amplitude random vibration with a strong first-mode component. For flow velocities less than critical, Paidoussis showed that the effect of the steady axial flow component is to damp free motions of the cylinders (Fig. 3). He proposed that the cause of the vibration was the cross-flow components that arise in real flows from nonuniformities (secondary circulation, separation behind the structure supporting the cylinder, and turbulence). The effects of cross-flow components of velocity are characterized by a force expression and a dissipation term. These were added to the free-vibration equation used in the stability study to obtain a more realistic equation for the response of a cylinder in an axial flow.

AMPLITUDE PREDICTIONS

Uncertainty in predicting the cross-flow component of velocity and the forcing function makes it impossible to predict from theory the amplitude of vibration; an empirical expression is needed. In his earlier paper[36] Paidoussis developed, using available experimental results, the empirical expression for amplitude given by Eq. 4. To account for his postulate (indirectly supported by experimental observations) that the amplitude of subcritical vibration is proportional to the departures from steady, purely axial flow, Paidoussis proposes a modified formula:

$$\frac{\delta}{D} = K\alpha^{-4}\left[\frac{(u^2 N_R \epsilon^2)^{4/3}}{1 + 2u^2}\right]\left[\frac{\beta^{2/3}}{1 + 4\beta}\right] \qquad (5)$$

where K is a constant depending primarily on the ratio of the cross-flow velocity to the axial flow velocity and is approximately equal to 10^{-6} for a system with minimum flow disturbance and 5×10^{-5} for highly disturbed flow conditions.

An experimental program is being conducted at the Chalk River Nuclear Laboratories (Atomic Energy of Canada Ltd.) to investigate systematically the effects of transverse flow, periodic fluctuations in the flow, and turbulence level on the amplitude of vibrations of cylinders in axial flow. Preliminary experiments using removable flow straighteners and acoustic filters have shown the dependence of vibration amplitude on flow disturbance (Fig. 4).

In addition to the general investigations of parallel-flow-induced vibrations, several experimental programs have been conducted in relation to specific reactor designs to define flow-induced vibration problems. Shields[27] reports on such tests in connection with the fuel rods of the N.S. *Savannah*, and Quinn[30] studied SEFOR fuel.

MECHANISMS

There is some disagreement among the various investigators whether parallel-flow vibration is forced

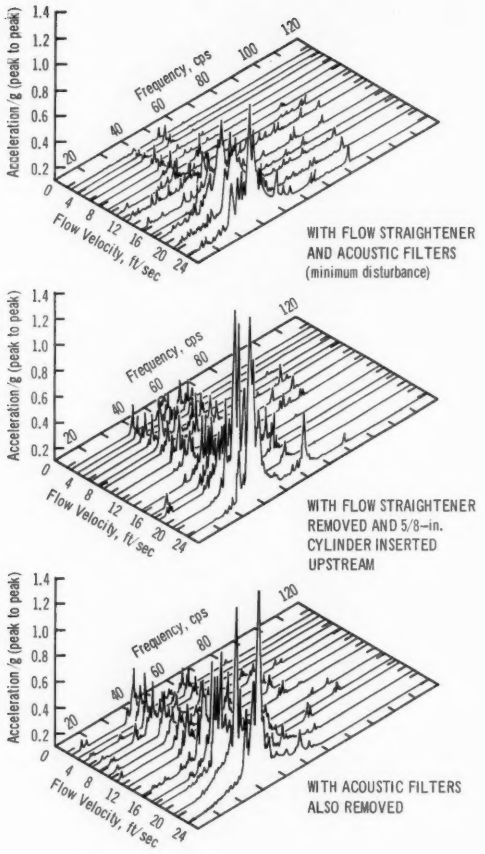

Acceleration /g (peak to peak)
Frequency, cps
Flow Velocity, ft/sec
WITH FLOW STRAIGHTENER AND ACOUSTIC FILTERS (minimum disturbance)
WITH FLOW STRAIGHTENER REMOVED AND 5/8-in. CYLINDER INSERTED UPSTREAM
WITH ACOUSTIC FILTERS ALSO REMOVED

Fig. 4 Vibration increases as flow disturbances increase.[37]

or self-excited. Burgreen,[26] Quinn,[28] and the Société Grenobloise d'Étude et d'Applications Hydrauliques[35] believe the mechanism is self-excitation. Foremost among their arguments is the fact that parallel-flow vibrations occur at all flow velocities with a frequency always near the natural frequency of vibration of the cylinder. On the other hand, Paidoussis[36,37] contends that the vibrations are force-excited by small, cross-flow velocity components.

Whether the parallel-flow vibrations are regarded as force-excited or self-excited depends on the viewpoint taken in identifying what is the system and what constitutes the environment. For example, consider the complete test section including the flowing fluid to be the system, and presuppose that the coolant possesses no alternating properties of its own and further that no energy is transmitted from the environment to the test section. Since there are no external forces present, the argument can be made that, for the present system description, any resulting vibrations are self-excited.

Alternatively, we might consider the cylinder to be the system, in which case the flowing fluid becomes part of the environment. If the cylinder, when fixed in its equilibrium position, experiences lateral forces, any resulting vibration would necessarily have to be termed force-excited. This is true if we are to accept the definition of self-excited vibrations, which states that the altering force sustaining the motion is created or controlled by the motion and disappears when the motion stops. Paidoussis contends that there are cross-flow components of velocity which do not arise from motions of the cylinder. These cross-flow components give rise to forces in the absence of cylinder motion, and therefore the resulting parallel-flow vibrations would be termed force-excited.

Flat-Plate Vibrations

The fuel in present-day power reactors is primarily in the configuration of fuel pins or fuel rods, rather than as plate-type elements. However, there is a limited interest in plate-type fuel elements for research reactors, and, in addition, the fluid-structure coupling in parallel-plate assemblies relates to the coupling of such reactor components as blade-type control rods.

DYNAMIC MODELS

High-velocity coolant flow through parallel-plate fuel assemblies can cause fuel-plate instability, plate divergence, and the more serious consequences of flow blockage and fuel melting. Miller[39] was the first to present a theoretical analysis that predicted the flow velocity at which collapse occurred. By applying a method of "neutral equilibrium," whereby pressure forces and plate restoring forces were balanced, he derived the following expression for the critical flow velocity, as a function of system parameters:

$$v_{CR} = \left[\alpha \frac{E_c{}^3 h}{(1-\nu^2)\rho_f l^4}\right]^{1/2} \tag{6}$$

where α = mode-dependent constant
E = Young's modulus
c = plate thickness
h = initial channel depth
ν = Poisson's ratio
ρ_f = mass density of coolant
l = characteristic length

Rosenberg and Youngdahl[40] later formulated a dynamic model to describe the motion of a fuel plate in a parallel-plate assembly. They solved the resulting eigenvalue problem and obtained the critical coolant-flow velocities of Miller. It should be noted that Rosenberg and Youngdahl also showed that consideration of fluid inertia, in the case of periodically supported plates, leads to a different interpretation of "critical" velocity.

Detailed Studies. More detailed theoretical studies related to the hydraulic instability of parallel-plate assemblies have been performed by Johansson[41] and Scavuzzo.[42] These studies considered the response problem (prediction of deflections). Johansson expanded on Miller's analysis by including the effects of fluid friction and flow redistribution. However, the pressure-drop term was again linearized. Large deflections using the nonlinear hydraulic loading term have been considered,[42] and the effects of fluid friction and flow redistribution have been included.[43]

Application. Core designers seeking a good estimate and understanding of the phenomenon of a critical flow velocity will find the results of these more detailed studies too complicated for application. For example, inclusion of fluid friction effects requires knowledge of entrance and exit losses and friction factors, quantities which are generally obtained empirically. As a further example, the end result of Scavuzzo's work is an integral equation that can be solved by an iterative procedure on the digital computer. Because of these complexities, and the results of analysis that show Miller's equations give a good indication of the coolant velocities at which plate motion can be expected, his results are frequently applied and referenced.[43]

In a recent study,[44] nonlinear terms of the second order were retained in a stability analysis of the equation of motion of a flat fuel plate in a reactor parallel-plate fuel assembly. The second-order terms generate an additional stability criterion in the form of an upper limit (critical deflection) on the amplitude of quasi-static deflections for stable oscillations, or neutral stability, about the undeflected fuel-plate position. The critical flow velocities derived in the earlier studies,[39,40] and again as a consequence of this analysis, are upper limits to ensure local stability about the undeflected plate position. A design constant, k, has been defined as

$$k = \frac{\delta_{cr}}{h} \tag{7}$$

where δ_{cr} is the critical deflection and h is the initial channel depth. New critical flow velocities have been defined. The new critical velocities ensure neutral stability about the undeflected plate position, for all steady-state and quasi-static displacements up to k (the design constant) times the initial channel depth. In terms of the original (Miller's) critical velocity (v_{CR}, from Eq. 6), the new critical velocity is given by

$$V_{cr} = \gamma v_{CR}$$

where $\gamma = [1 + (k/\beta)^2]^{-1/2}$ and β depends on the edge fixity. The coefficient γ is plotted as a function of k for various boundary conditions in Fig. 5. From Fig 5 it can be seen that for $k = 0.5$ (the maximum physically realizable value as governed by the assumptions), the new critical flow velocities vary from 0.633 to 0.852 times those first derived by Miller.

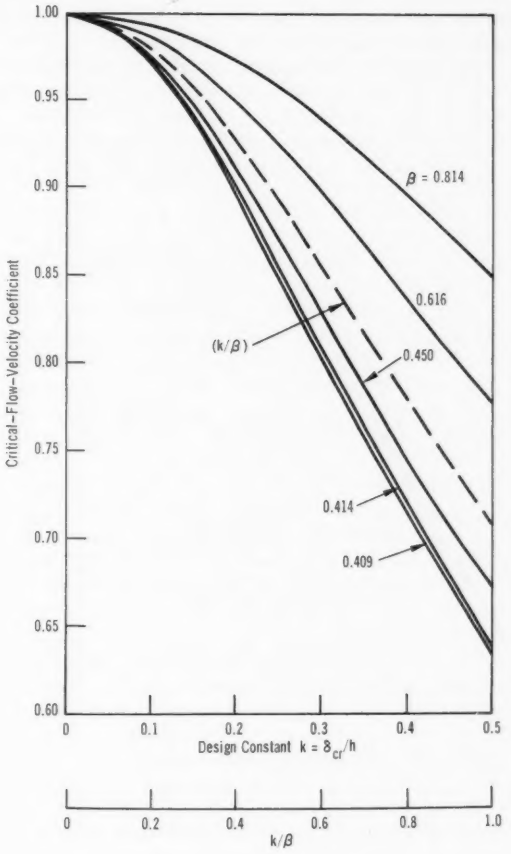


Fig. 5 Boundary conditions influence the design constant k *and the critical-flow-velocity coefficient* γ.[44]

Blade Behavior. Other papers[45,46] present a theoretical analysis of flow-induced vibration of a blade suspended in a flow channel. Two modes of motion, translational and angular, are analyzed independently. The physical situation is that of a blade suspended in a flow channel which widens at a point downstream of the leading edge of the blade.

Neglecting cross flow from one side of the blade to the other, the stability problem is solved to give the boundaries of the three regions of possible motion, namely, decaying vibrations, divergent vibrations, and hydraulic locking to the channel wall (a static type of divergence). The three regions of motion are represented in Fig. 6 as a function of flow rate and length of insertion of the blade into the narrow part (scabbard) of the flow channel. The solid line in Fig. 6 that separates the regions of divergent and decaying vibrations corresponds to a net damping of zero and gives

the flow rate at which infinitesimal-constant-amplitude oscillations are sustained.

Qualitatively, the results explain the behavior observed in an experimental water table model. The results indicate measures that may be effective in correcting a vibration problem on a suspended blade or other bodies caused to vibrate by the same phenomenon.

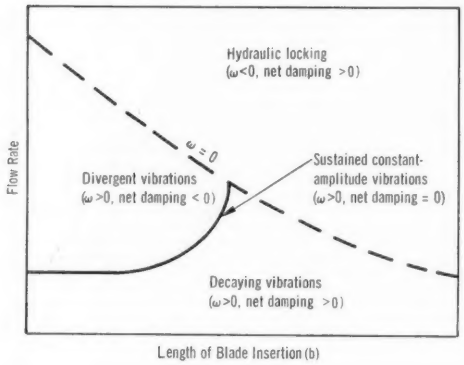


Fig. 6 Vibration of a blade in a channel can be increasing, steady, or decreasing; in the extreme the divergent blade is locked against a channel wall.[46]

Damping, Modeling, and Scaling

We are interested in the amplitudes of stable vibrations because small displacements in a reactor can have serious effects; the response problem must be considered. To determine the amplitude of a vibration from analytical considerations alone, we must know the energy dissipative mechanisms and particularly the governing damping laws.

DAMPING STUDIES

A method of identifying and evaluating linear damping models in beam vibration resulted from a study[47] of a simulated reactor core that consisted of a cluster of hexagonal rods clamped together at the base. The inherent material damping was assumed neligible when compared to the damping due to interaction among the clustered rods and damping at the joint. To model this interaction damping mathematically, we assumed three damping mechanisms. A theoretical analysis, using the Euler beam theory and assuming the damping is sufficiently small that natural frequencies and mode shapes are unaffected, gives the ratio of first- to second-mode magnification factors as constants corresponding to "pure forms" for each of the proposed damping mechanisms.

Identification of the dominant damping mechanisms for a particular physical model involves the experimental determination of the ratio of magnification factors (related to a section of the beam) and comparison of these ratios with the corresponding ratio constants determined from theoretical considerations. The associated damping coefficients are found by solving a set of independent simultaneous equations (constructed by expressing the coefficients as a linear combination and using experimental results obtained at resonant conditions).

The experimental studies in this program were conducted with atmospheric air as the fluid environment. In reactors the core components are surrounded by a liquid (water or sodium) environment that can be expected to have a significant effect on damping. Keane studied the force vibration of a cantilever tube in air and water, both analytically and by experiment.[48] However, he was primarily concerned with the accession to inertia phenomenon and did not directly treat the damping problem.

STRUCTURAL MODELING

The linear response of a structure to an arbitrary dynamic excitation can be represented as a superposition, or linear combination, of normal modes. This points out the importance of determining the eigenvalues and eigenfunctions of reactor core structures. Toward this end, Bohm[49] approximated the reactor internals by an interconnected beam-type structure and presented a transfer-matrix approach for the determination of natural frequencies and normal modes. Results for two cases are presented: (1) for the reactor internals with a rigid vessel support and (2) for the internals supported by an elastic vessel. The structural models are shown in Fig. 7. The results for reactor internals supported by an elastic vessel are given in Fig. 8. The models studied were highly idealized. Nevertheless, the basic method of analysis (representing the response as a superposition of normal modes) has been proven (for linear systems), and the approach lays the foundation upon which the analyses of more complex structures can be built, for example, coupled fuel-element and grid components.

A nonlinear problem has been reported[50,51] in which structures (fuel elements) with relatively small clearances between adjacent components were subjected to oscillating excitation. This caused the system components to be impinged upon one another. This study also included the case of a vertical beam that was subjected to a forced vibration at the upper end, and, when its deflections exceeded a prescribed magnitude, it was hitting an elastic wall at the lower end. The dynamic response problems was solved by numerical integration of the differential equation of motion using a digital analog simulator. The elastic wall was characterized by a number of springs with nonlinear spring characteristics.

At this time dynamic response cannot be predicted from theory with sufficient accuracy. Although useful

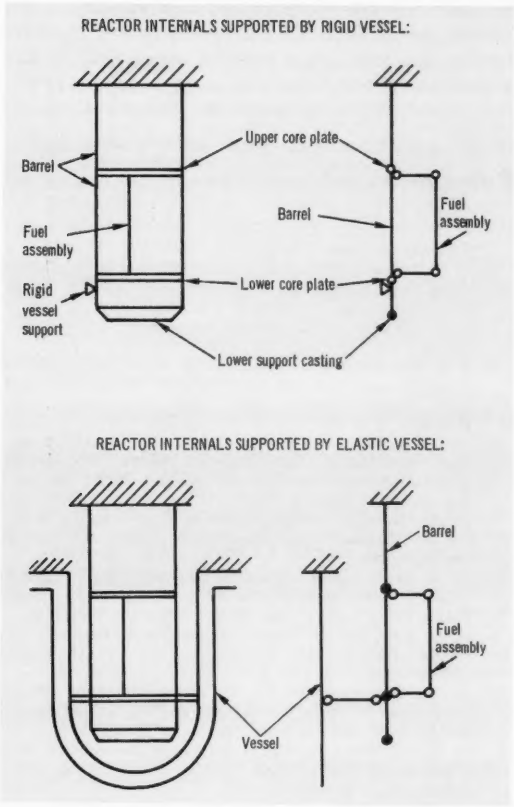


Fig. 7 Two models of reactor internals: the upper one provides rigid support for the core barrel and the lower, an elastic vessel.[49]

estimates of resonant and unstable conditions can be obtained, only prototype testing and/or the ultimate test of the as-built design establish the vibrational characteristics of core internals.

SCALING EXPERIMENTS

As discussed earlier, it is not always acceptable economically and can be unsafe to depend on the operation of the reactor to indicate unknown vibrational problems related to the design. Keane recognized the need to develop an improved approach to vibrational problems encountered in reactor design.[52] As part of a program undertaken to investigate the feasibility of dynamic model testing of reactor components, a core thermocouple support tube was studied. A dynamic scale model was fabricated using model laws derived from the equation for free vibration of a beam. A comparison of the response of model and prototype to forced vibration is given. The results were good and justify further investigations in the field of dynamic testing of core components.

The testing of a full-size core mockup is often prohibitive due primarily to the high cost of fabricating prototype fuel subassemblies. Therefore it would be desirable to obtain information regarding experiment scaling in the sense of the number of elements required to reveal the mechanical characteristics of the full prototype complement of subassemblies. Preliminary information can be inferred from the results of damping-model studies.[47] Eight cluster sizes, consisting of 1, 7, 19, 37, 61, 91, 127, and 163 rods, simulating fuel assemblies of a reactor core, were tested. The test results showed the ratio of first- to second-mode magnification factors to be nearly invariant with cluster size. Tests also

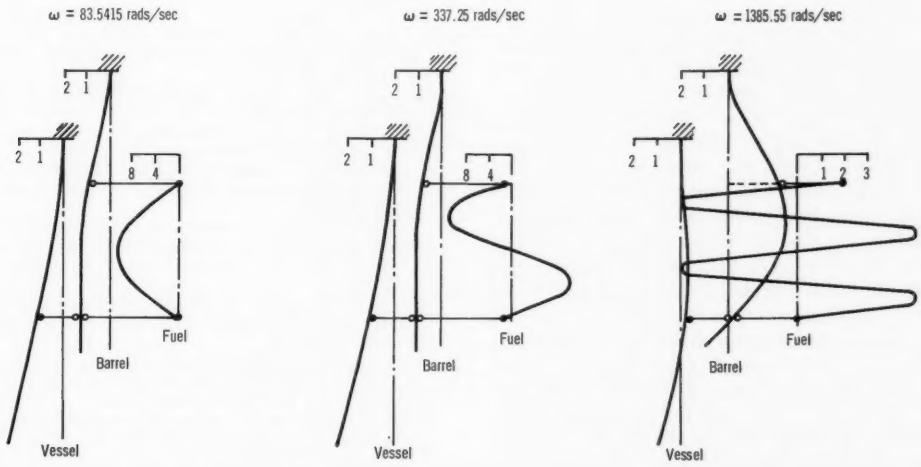


Fig. 8 Natural frequencies and normal modes of vibration for reactor internals supported by an elastic vessel.[49]

showed that the magnification factors asymptotically approached constant values as cluster size increased. With such information a small number of prototype components can be tested (for example, at first mode) and the results extrapolated to obtain characteristic behaviors of a full-size complement.

In Brief

The vibration of reactor core components is greatly complicated by the flowing-coolant environment and presents unique problems to the dynamicist.

The stability problem and resonant phenomena have received considerable attention. Within the framework of the assumptions of the respective analyses, the stability problem may be considered solved at least for idealized models and systems closely approximating them.

The fact that small clearances are involved in reactors, say between adjacent fuel pins, and that changes in these clearances can result in power oscillations[1] or have more serious consequences should loss of coolant result, leading to fuel melting, makes *small displacements critical.* It is this criticalness of small displacements that contributes to making the response problem—the prediction of vibration amplitudes—important to core designers.

The response of parallel-plate assemblies has received some consideration.[41-43] However, in sodium-cooled fast reactors, the fuel generally takes the configuration of pins or rods rather than plates. Toward this end (the prediction of the amplitude of parallel-flow-induced vibration in rods), four empirical expressions have been proposed[26,28,36,37] However, even the most successful of the four, that of Paidoussis,[37] does not account for several features of the system—for example, a characterization of the forcing function, dissipation of energy, and a temperature effect. The primary reason that these parameters are not included in the empirical expression is that few experimental data were available in which these parameters were varied.

The state-of-the-art survey indicates the need for basic research related to the phenomena of flow-induced vibration of rods and plates, before considering the complex geometries and boundary conditions of reactor core components. This basic research should involve studies of the damping and energy dissipation of a rod and rod bundles in a flowing liquid and the development of appropriate damping laws. Also, because the flow-induced response is a random vibration, methods of analyzing stochastic systems[53] and of characterizing the random forcing function, or excitation, must be studied and new methods developed where necessary. A random vibration approach has been identified[54] and is currently being applied to predict mean-square response.

The complexity of the response problem seems to preclude the possibility of obtaining a complete analytic relation. However, an understanding of the various mechanisms associated with flow-induced vibration may give design insights, which, coupled with simple experimental measurements made on a prototype model, will allow meaningful design evaluations.

References

1. R. W. Hyndman et al., EBR-II Self-Excited Oscillations, *Trans. Amer. Nucl. Soc.*, 8(2): 500 (1965).
2. C. A. Dalke, Elimination of Hydraulically Induced Core Vibrations in the General Electric Test Reactor, *Trans. Amer. Nucl. Soc.*, 8(Suppl.): 2 (1965).
3. R. C. Skaardal, Flow-Induced Vibrations, *Power Reactor Technol.*,8(4): 273-275 (Fall 1965).
4. J. I. Riesland and E. A. Gustafson, Work Performed on Fuel Channels and the Core Support Plate at Big Rock Point Nuclear Power Plant, *Trans. Amer. Nucl. Soc.*, 8(Suppl.): 5-9 (1965).
5. Action on Reactor Projects Undergoing Regulatory Review, *Nucl. Safety*, 6(2): 222-223 (Winter 1964–1965).
6. Thermal Shield Vibration at Big Rock Point, *Nucleonics*, 24(5): 64-65 (May 1966).
7. F. H. Martens, Design Foresight Can Simplify Reactor Modification, *Power Reactor Technol.*, 9(2): 74-79 (Spring 1966).
8. G. A. Reed, Major Operating and Refueling Problems at the Yankee Atomic Electric Company Rowe Power Plant, *Trans. Amer. Nucl. Soc.*, 8(Suppl.): 28 (1965).
9. G. A. Reed and E. Tarnuzzer, Examining Yankee Plant Performance in 1965, *Nucleonics*, 24(3): 42-47 (March 1966).
10. G. F. Bierman and W. J. Miller, Nuclear Plant Performance—Good and Getting Better, *Power Reactor Technol.*, 9(3): 110-122 (Summer 1966).
11. R. E. Heffner, Six-Years Operating Experience with PWR Plant Components, *Trans. Amer. Nucl. Soc.*, 8(Suppl.): 29-30 (1965).
12. Oak Ridge National Laboratory, Molten-Salt Reactor Program Semiannual Progress Report for Period Ending January 31, 1964, USAEC Report ORNL-3626, July 1964.
13. Oak Ridge National Laboratory, Molten-Salt Reactor Program Semiannual Progress Report for Period Ending July 31, 1964, USAEC Report ORNL-3708, November 1965.
14. W. K. Stromquist and O. Sisman, High Flux Reactor Fuel Assembly—Vibration and Water Flow, USAEC Report ORNL-50, Oak Ridge National Laboratory, June 15, 1948.
15. Y. C. Fung, *The Theory of Aeroelasticity*, p. 67, John Wiley & Sons, Inc., New York, 1955.
16. P. Leon, Study of Vibrations and Pressure Losses in Nests of Tubes, Preliminary Theoretical Study, Special Report No. 1, USAEC–Euratom Report EURAEC-160, Société Grenobloise d'Étude et d'Applications Hydrauliques, 1961.
17. A. W. Harris, A Review on Vortex Sheets, Periodic Wakes, and Induced Vibration Phenomena, *Trans. ASME (Amer. Soc. Mech. Eng.), Ser. D, J. Basic Eng.*, 86: 185 (1964).
18. J. L. Livesay and R. C. Dye, Vortex Excited Vibration of a Heat Exchanger Tube Row, *J. Mech. Eng. Sci.*, 4(4): 349 (1962).
19. A. A. Putnam, Flow Induced Noise in Heat Exchangers, *Trans. ASME (Amer. Soc. Mech. Eng.), Ser. A, J. Eng. Power*, 81: 417-422 (October 1959).
20. B. J. Grotz and E. R. Arnold, Flow Induced Vibrations in Heat Exchangers, Report AD-104568, Stanford University, 1956.
21. J. P. DenHartog, *Mechanical Vibrations*, p. 307, McGraw-Hill Book Company, Inc., New York, 1956.
22. H. Ashley and G. Haviland, Bending Vibrations of a Pipe-

line Containing Flowing Fluid, *Trans. ASME (Amer. Soc. Mech. Eng.), Ser. E, J. Appl. Mech.*, 72: 229-232 (1950).
23. G. W. Hausner, Bending Vibrations of a Pipeline Containing Flowing Fluid, *Trans. ASME (Amer. Soc. Mech. Eng.). Ser. E, J. Appl. Mech.*, 74: 205-208 (1952).
24. F. I. Nioidson, Vibrations of a Cylindrical Tube Containing Flowing Fluid, *Trans. Roy. Inst. Technol., Stockholm*, 73: (1953).
25. R. H. Long, Experimental and Theoretical Studies of Transverse Vibration of a Tube Containing Flowing Fluid, *Trans. ASME (Amer. Soc. Mech. Eng.), Ser. E, J. Appl. Mech.*, 77: 65-68 (1955).
26. D. Burgreen, J. J. Byrnes, and D. M. Beneforado, Vibration of Rods Induced by Water in Parallel Flow, *Trans. Amer. Soc. Mech. Eng.*, 80: 991 (July 1958).
27. C. M. Shields, N. S. Savannah Fuel Design and Development Program — Fuel Rod Vibration, USAEC Report GEAP-3583, General Electric Company, Atomic Power Equipment Department, Nov. 1, 1960.
28. E. P. Quinn, Vibration of Fuel Rods in Parallel Flow, USAEC Report GEAP-4059, General Electric Company, Atomic Power Equipment Department, July 1962.
29. R. T. Pavlica and R. C. Marshall, Vibration of Fuel Assemblies in Parallel Flow, *Trans. Amer. Nucl. Soc.*, 8(2): 599-600 (1965).
30. E. P. Quinn, Vibration of SEFOR Fuel Rods in Parallel Flow, USAEC Report GEAP-4966, General Electric Company, Atomic Power Equipment Department, September 1965.
31. R. T. Pavlica and R. C. Marshall, An Experimental Study of Fuel Assembly Vibrations Induced by Coolant Flow, *Nucl. Eng. Design*, 4: 54-60 (1966).
32. K. G. Roström and N. *Andersson*, Boiler Element for Marviken, Vibration Tests with One Rod, Swedish Report RPL-724, 1964.
33. K. G. Roström, Super Heater Element for Marviken, Vibration Test with One Rod, Swedish Report RPL-725, 1964.
34. K. G. Roström, Seven-Rod Fuel Element Vibration Test, Swedish Report RPL-726, 1964.
35. Société Grenobloise d'Etude et d'Applications Hydrauliques, Study of Vibrations and Load Losses in Tubular Clusters, Special Report No. 3, USAEC – Euratom Report EURAEC-288, March 1962.
36. M. P. Paidoussis, The Amplitude of Fluid-Induced Vibrations of Cylinders in Axial Flow, Canadian Report AECL-2225, March 1965.
37. M. P. Paidoussis, Vibration of Flexible Cylinders with Supported Ends, Induced by Axial Flow, *Proc. Inst. Mech. Engrs.*, 180(Pt. 3J): (1965–1966).
38. M. P. Paidoussis, Dynamics of Flexible Slender Cylinders in Axial Flow. Part 1, Theory, *J. Fluid Mech.*, 26(Pt. 4): 717-736 (December 1966).
39. D. R. Miller, Critical Flow Velocities for Collapse of Reactor Parallel-Plate Fuel Assemblies, *J. Eng. Power*, 82: 83-95 (1960).
40. C. S. Rosenberg and C. K. Youngdahl, A Simplified Dynamic Model for the Vibration Frequencies and Critical Coolant Flow Velocities for Reactor Parallel Fuel Assemblies, *Nucl. Sci. Eng.*, 13: 91-102 (1962).
41. E. B. Johansson, Hydraulic Instability of Reactor Parallel-Plate Fuel Assemblies, USAEC Report KAPL-M-EJ-9, Knolls Atomic Power Laboratory, July 1959.
42. R. J. Scavuzzo, Hydraulic Instability of Flat Parallel-Plate Assemblies, *Nucl. Sci. Eng.*, 21: 463-472 (1965).
43. R. J. Scavuzzo, Hydraulic Instability of Flat Parallel-Plate Assemblies, Ph. D. Dissertation, University of Pittsburgh, 1962.
44. M. W. Wambsganss, Second Order Effects as Related to Critical Coolant Flow Velocities and Reactor Parallel Plate Fuel Assemblies, *Nucl. Eng. Design*, 5(3): 268-276 (1967); also, USAEC Report ANL-7261, Argonne National Laboratory, November 1966.
45. D. R. Miller and R. G. Kennison, Theoretical Analysis of Flow-Induced Vibration of a Blade Suspended in a Flow Channel, presented at Winter Annual Meeting of ASME, New York, Nov. 27 – Dec. 1, 1966, Paper No. 66-WA/NE-1.
46. M. W. Wambsganss, Flow-Induced Vibration in Reactor Internals, *Power Reactor Technol. and Reactor Fuel Process.*, 10(1): 2-5 (Winter 1966–1967).
47. M. W. Wambsganss, B. L. Boers, and G. S. Rosenberg, A Method for Identifying and Evaluating Linear Damping Models in Beam Vibrations, *Shock Vib. Bull.*, 36(4): 65-75 (1967); also, USAEC Report ANL-7292, Argonne National Laboratory, April 1967.
48. J. A. Keane, Forced Vibration of a Cantilever Tube in Air and Water, in Bettis Technical Review — Reactor Technology, USAEC Report WAPD-BT-18, pp. 75-84, Westinghouse Electric Corp., Bettis Atomic Power Laboratory, April 1960.
49. G. J. Bohm, Natural Vibration of Reactor Internals, *Nucl. Sci. Eng.*, 22: 143-152 (1965).
50. A. N. Nahavandi and G. J. Bohm, A Solution of Nonlinear Vibration Problems in Reactor Components, *Trans. Amer. Nucl. Soc.*, 8(2): 491-492 (1965).
51. A. N. Nahavandi and G. J. Bohm, A Solution of Nonlinear Vibration Problems in Reactor Components, *Nucl. Sci. Eng.*, 26: 80-89 (1966).
52. J. A. Keane, A Feasibility Study of Model Testing To Establish the Vibrational Integrity of Reactor Core Components, *Trans. Amer. Nucl. Soc.*, 4(1): 63-64 (1961).
53. Y. K. Lin, *Probabilistic Theory of Structural Dynamics*, McGraw-Hill Book Company, Inc., New York, 1967.
54. Argonne National Laboratory, Core Structural Dynamics, in Reactor Development Program Progress Report for February 1967, USAEC Report ANL-7308, p. 69, March 1967.

AEC Revises General Design Criteria for Nuclear Power Plants

As part of the nuclear industry efforts to evolve improved reactor design and safety standards based on experience and improved technology, the U. S. Atomic Energy Commission (AEC) on July 11, 1967, published a revised set of 70 "General Design Criteria for Nuclear Power Plant Construction Permits" in the *Federal Register*. The criteria are intended to help a prospective applicant for a construction permit establish the principal design safety requirements for his plant. They reflect AEC interest in defining codes and standards to assure safe designs by means of engineering safeguards features, redundancy and independence of key components and emergency power supplies, quality-assurance procedures, and continuing inspectability of components during plant life.

The Revised Version. The Commission published its first list of 27 proposed criteria on Nov. 22, 1965, requesting comments. Compiled by the regulatory staff, the criteria stated reactor-plant design principles and objectives that had evolved over the years. The more detailed list published in July, for interim use and for further comments by the industry, reflects extensive suggestions made by 20 groups or individuals, a task force of the Atomic Industrial Forum's committee on reactor safety, the Commission's Advisory Committee on Reactor Safeguards, and reviews within the AEC.

Further Changes. Although plant designers, construction-permit applicants, and the AEC regulatory staff will use the July 11, 1967, list for guidance, the list is still subject to further revision after the Commission considers comments received by Sept. 9, 1967. In time the final version will be published in the *Code of Federal Regulations*, Appendix A to the AEC's 10 CFR Part 50, Licensing of Production and Utilization Facilities.

How They Are To Be Used. Part 50.34 of the AEC's regulations requires that every application for a construction permit include the principal design criteria for the proposed plant. The general design criteria are intended as a guide for formulating and judging these. Although the general criteria reflect mostly the experience acquired with pressurized-water- and boiling-water-reactor plants as designed and sited so far, the Commission regards them as being generally applicable to all types of power reactors. However, the AEC regulations also require the applicant to assure that his principal design criteria encompass all plant design features needed for public health and safety. Thus there may be some specific plants, types of reactors, sites, or environmental conditions for which the principal design criteria should justifiably differ from the general criteria.

On the basis of its experience with the licensing procedure, the Commission has found that, at the construction-permit stage, applicants need to furnish more definitive information for some criteria than for others that can be considered further later. The 47 design features designated as Category A items (see superscripts after criteria titles) are those which need to be considered in the most detail for a construction permit. Category B items mostly concern control and monitoring systems, fuel and waste storage, and plant effluent.

The 70 criteria, organized in nine groups, follow.

I Overall Plant Requirements

1. *Quality Standards.*[A] Those systems and components of reactor facilities which are essential to the prevention of accidents that could affect the public health and safety or to mitigation of their consequences shall be identified and then designed, fabricated, and erected to quality standards that reflect the importance of the safety function to be performed. Where generally recognized codes or standards on design, materials, fabrication, and inspection are used, they shall be identified. Where adherence to such codes or standards does not suffice to assure a quality product in keeping with the safety function, they shall be supplemented or modified as necessary. Quality-assurance programs, test procedures, and inspection acceptance levels to be used shall be identified. A showing of sufficiency and

applicability of codes, standards, quality-assurance programs, test procedures, and inspection acceptance levels used is required.

2. *Performance Standards.*[A] Those systems and components of reactor facilities which are essential to the prevention of accidents that could affect the public health and safety or to mitigation of their consequences shall be designed, fabricated, and erected to performance standards that will enable the facility to withstand, without loss of the capability to protect the public, the additional forces that might be imposed by natural phenomena such as earthquakes, tornadoes, flooding conditions, winds, ice, and other local site effects. The design bases so established shall reflect: (1) appropriate consideration of the most severe of these natural phenomena which have been recorded for the site and the surrounding area and (2) an appropriate margin for withstanding forces greater than those recorded to reflect uncertainties about the historical data and their suitability as a basis for design.

3. *Fire Protection.*[A] The reactor facility shall be designed (1) to minimize the probability of events such as fires and explosions and (2) to minimize the potential effects of such events to safety. Noncombustible and fire-resistant materials shall be used whenever practical throughout the facility, particularly in areas containing critical portions of the facility such as containment, control room, and components of engineered safety features.

4. *Sharing of Systems.*[A] Reactor facilities shall not share systems or components unless it is shown that safety is not impaired by the sharing.

5. *Records Requirements.*[A] Records of the design, fabrication, and construction of essential components of the plant shall be maintained by the reactor operator or under his control throughout the life of the reactor.

II Protection by Multiple Fission-Product Barriers

6. *Reactor Core Design.*[A] The reactor core shall be designed to function throughout its design lifetime, without exceeding acceptable fuel-damage limits that have been stipulated and justified. The core design, together with reliable process and decay-heat removal systems, shall provide for this capability under all expected conditions of normal operation, with appropriate margins for uncertainties and for transient situations that can be anticipated, including the effects of the loss of power to recirculation pumps, tripping out of a turbine-generator set, isolation of the reactor from its primary heat sink, and loss of all off-site power.

7. *Suppression of Power Oscillations.*[B] The core design, together with reliable controls, shall ensure that power oscillations that could damage in excess of acceptable fuel-damage limits are not possible or can be readily suppressed.

8. *Overall Power Coefficient.*[B] The reactor shall be designed so that the overall power coefficient in the power operating range shall not be positive.

9. *Reactor Coolant-Pressure Boundary.*[A] The reactor coolant-pressure boundary shall be designed and constructed so as to have an exceedingly low probability of gross rupture or significant leakage throughout its design lifetime.

10. *Containment.*[A] Containment shall be provided. The containment structure shall be designed to sustain the initial effects of gross equipment failures, such as a large coolant-boundary break, without loss of required integrity and, together with other engineered safety features as may be necessary, to retain for as long as the situation requires the functional capability to protect the public.

III Nuclear and Radiation Controls

11. *Control Room.*[B] The facility shall be provided with a control room from which actions to maintain safe operational status of the plant can be controlled. Adequate radiation protection shall be provided to permit access, even under accident conditions, to equipment in the control room or other areas as necessary to shut down and maintain safe control of the facility without radiation exposures of personnel in excess of 10 CFR 20 limits. It shall be possible to shut the reactor down and maintain it in a safe condition if access to the control room is lost due to fire or other cause.

12. *Instrumentation and Control Systems.*[B] Instrumentation and controls shall be provided as required to monitor and maintain variables within prescribed operating ranges.

13. *Fission-Process Monitors and Controls.*[B] Means shall be provided for monitoring and maintaining control over the fission process throughout core life and for all conditions that can reasonably be anticipated to cause variations in reactivity of the core, such as indication of position of control rods and concentration of soluble reactivity-control poisons.

14. *Core Protection Systems.*[B] Core protection systems, together with associated equipment, shall be designed to act automatically to prevent or to suppress conditions that could result in exceeding acceptable fuel-damage limits.

15. *Engineered-Safety-Features Protection Systems.*[B] Protection systems shall be provided for sensing accident situations and initiating the operation of necessary engineered safety features.

16. *Monitoring Reactor Coolant-Pressure Boundary.*[B] Means shall be provided for monitoring the reactor coolant-pressure boundary to detect leakage.

17. *Monitoring Radioactivity Releases.*[B] Means shall be provided for monitoring the containment

atmosphere, the facility effluent-discharge paths, and the facility environs for radioactivity that could be released from normal operations, from anticipated transients, and from accident conditions.

18. *Monitoring Fuel and Waste Storage.*[B] Monitoring and alarm instrumentation shall be provided for fuel and waste storage and handling areas for conditions that might contribute to loss of continuity in decay-heat removal and to radiation exposures.

IV Reliability and Testability of Protection Systems

19. *Protection-Systems Reliability.*[B] Protection systems shall be designed for high functional reliability and in-service testability commensurate with the safety functions to be performed.

20. *Protection-Systems Redundancy and Independence.*[B] Redundancy and independence designed into protection systems shall be sufficient to assure that no single failure or removal from service of any component or channel of a system will result in loss of the protection function. The redundancy provided shall include, as a minimum, two channels of protection for each protection function to be served. Different principles shall be used where necessary to achieve true independence of redundant instrumentation components.

21. *Single Failure Definition.*[B] Multiple failures resulting from a single event shall be treated as a single failure.

22. *Separation of Protection and Control Instrumentation Systems.*[B] Protection systems shall be separated from control instrumentation systems to the extent that failure or removal from service of any control instrumentation system component or channel, or of those common to control instrumentation and protection circuitry, leaves intact a system satisfying all requirements for the protection channels.

23. *Protection Against Multiple Disability for Protection Systems.*[B] The effects of adverse conditions to which redundant channels or protection systems might be exposed in common, either under normal conditions or those of an accident, shall not result in loss of the protection function.

24. *Emergency Power for Protection Systems.*[B] In the event of loss of all off-site power, sufficient alternate sources of power shall be provided to permit the required functioning of the protection systems.

25. *Demonstration of Functional Operability of Protection Systems.*[B] Means shall be included for testing protection systems while the reactor is in operation to demonstrate that no failure or loss of redundancy has occurred.

26. *Protection-Systems Fail-Safe Design.*[B] The protection systems shall be designed to fail into a safe state or into a state established as tolerable on a defined basis if such conditions as disconnection of the system, loss of energy (e.g., electric power and instrument air), or adverse environments (e.g., extreme heat or cold, fire, steam, or water) are experienced.

V Reactivity Control

27. *Redundancy of Reactivity Control.*[A] At least two independent reactivity-control systems, preferably of different principles, shall be provided.

28. *Reactivity Hot-Shutdown Capability.*[A] At least two of the reactivity-control systems provided shall independently be capable of making and holding the core subcritical from any hot standby or hot operating condition, including those resulting from power changes, sufficiently fast to prevent exceeding acceptable fuel-damage limits.

29. *Reactivity Shutdown Capability.*[A] At least one of the reactivity-control systems provided shall be capable of making the core subcritical under any condition (including anticipated operational transients) sufficiently fast to prevent exceeding acceptable fuel-damage limits. Shutdown margins greater than the maximum worth of the most effective control rod when fully withdrawn shall be provided.

30. *Reactivity Hold-Down Capability.*[B] At least one of the reactivity-control systems provided shall be capable of making and holding the core subcritical under any conditions with appropriate margins for contingencies.

31. *Reactivity-Control-Systems Malfunction.*[B] The reactivity-control systems shall be capable of sustaining any single malfunction, such as unplanned continuous withdrawal (not ejection) of a control rod, without causing a reactivity transient that could result in exceeding acceptable fuel-damage limits.

32. *Maximum Reactivity Worth of Control Rods.*[A] Limits, which include considerable margin, shall be placed on the maximum reactivity worth of control rods or elements and on rates at which reactivity can be increased to ensure that the potential effects of a sudden or large change of reactivity cannot (1) rupture the reactor coolant-pressure boundary or (2) disrupt the core, its support structures, or other vessel internals sufficiently to impair the effectiveness of emergency core cooling.

VI Reactor Coolant-Pressure Boundary

33. *Reactor Coolant-Pressure-Boundary Capability.*[A] The reactor coolant-pressure boundary shall be capable of accommodating without rupture, and with only limited allowance for energy absorption through plastic deformation, the static and dynamic loads imposed on any boundary component as a result of any inadvertent and sudden release of energy to the coolant. As a design reference, this sudden release shall be taken as that which would result

from a sudden reactivity insertion such as rod ejection (unless prevented by positive mechanical means), rod dropout, or cold-water addition.

34. *Reactor Coolant-Pressure-Boundary Rapid Propagation-Failure Prevention.*[A] The reactor coolant-pressure boundary shall be designed to minimize the probability of rapid propagating-type failures. Consideration shall be given (1) to the notch-toughness properties of materials extending to the upper shelf of the Charpy transition curve, (2) to the state of stress of materials under static and transient loadings, (3) to the quality control specified for materials and component fabrication to limit flaw sizes, and (4) to the provisions for control over service temperature and irradiation effects that may require operational restrictions.

35. *Reactor Coolant-Pressure-Boundary Brittle-Fracture Prevention.*[A] Under conditions where reactor coolant-pressure-boundary system components constructed of ferritic materials may be subjected to potential loadings, such as a reactivity-induced loading, service temperatures shall be at least 120° F above the nil ductility transition (NDT) temperature of the component material if the resulting energy release is expected to be absorbed by plastic deformation or 60° F above the NDT temperature of the component material if the resulting energy release is expected to be absorbed within the elastic strain-energy range.

36. *Reactor Coolant-Pressure-Boundary Surveillance.*[A] Reactor coolant-pressure-boundary components shall have provisions for inspection, testing, and surveillance by appropriate means to assess the structural and leaktight integrity of the boundary components during their service lifetime. For the reactor vessel a material surveillance program conforming with ASTM-E-185-66 shall be provided.

VII Engineered Safety Features

37. *Engineered Safety Features As Basis for Design.*[A] Engineered safety features shall be provided in the facility to back up the safety provided by the core design, the reactor coolant-pressure boundary, and their protection systems. As a minimum, such engineered safety features shall be designed to cope with any size reactor coolant-pressure-boundary break up to and including the circumferential rupture of any pipe in that boundary assuming unobstructed discharge from both ends.

38. *Reliability and Testability of Engineered Safety Features.*[A] All engineered safety features shall be designed to provide high functional reliability and ready testability. In determining the suitability of a facility for a proposed site, the degree of reliance upon and acceptance of the inherent and engineered safety afforded by the systems, including engineered safety features, will be influenced by the known and the demonstrated performance capability and reliability of the systems and by the extent to which the operability of such systems can be tested and inspected where appropriate during the life of the plant.

39. *Emergency Power for Engineered Safety Features.*[A] Alternate power systems shall be provided and designed with adequate independency, redundancy, capacity, and testability to permit the functioning required of the engineered safety features. As a minimum, the on-site power system and the off-site power system shall each, independently, provide this capacity assuming a failure of a single active component in each power system.

40. *Missile Protection.*[A] Protection for engineered safety features shall be provided against dynamic effects and missiles that might result from plant equipment failures.

41. *Engineered-Safety-Features Performance Capability.*[A] Engineered safety features such as emergency core-cooling and containment heat-removal systems shall provide sufficient performance capability to accommodate partial loss of installed capacity and still fulfill the required safety function. As a minimum, each engineered safety feature shall provide this required safety function assuming a failure of a single active component.

42. *Engineered-Safety-Features Components Capability.*[A] Engineered safety features shall be designed so that the capability of each component and system to perform its required function is not impaired by the effects of a loss-of-coolant accident.

43. *Accident-Aggravation Prevention.*[A] Engineered safety features shall be designed so that any action of the engineered safety features that might accentuate the adverse aftereffects of the loss of normal cooling is avoided.

44. *Emergency Core-Cooling-Systems Capability.*[A] At least two emergency core-cooling systems, preferably of different design principles, each with a capability for accomplishing abundant emergency core cooling, shall be provided. Each emergency core-cooling system and the core shall be designed to prevent fuel and cladding damage that would interfere with the emergency core-cooling function and to limit the cladding metal–water reaction to negligible amounts for all sizes of breaks in the reactor coolant-pressure boundary, including the double-ended rupture of the largest pipe. The performance of each emergency core-cooling system shall be evaluated conservatively in each area of uncertainty. The systems shall not share active components and shall not share other features or components unless it can be demonstrated that (1) the capability of the shared feature or component to perform its required function can be readily ascertained during reactor operation, (2) failure of the shared feature or component does not initiate a loss-of-coolant accident, and (3) capability of the shared feature or component to perform its required function is not impaired by

the effects of a loss-of-coolant accident and is not lost during the entire period this function is required following the accident.

45. *Inspection of Emergency Core-Cooling Systems.*[A] Design provisions shall be made to facilitate physical inspection of all critical parts of the emergency core-cooling systems, including reactor-vessel internals and water-injection nozzles.

46. *Testing of Emergency Core-Cooling-Systems Components.*[A] Design provisions shall be made so that active components of the emergency core-cooling systems, such as pumps and valves, can be tested periodically for operability and required functional performance.

47. *Testing of Emergency Core-Cooling Systems.*[A] A capability shall be provided to test periodically the delivery capability of the emergency core-cooling systems at a location as close to the core as is practical.

48. *Testing of Operational Sequence of Emergency Core-Cooling Systems.*[A] A capability shall be provided to test under conditions as close to design as practical the full operational sequence that would bring the emergency core-cooling systems into action, including the transfer to alternate power sources.

49. *Containment Design Basis.*[A] The containment structure, including access openings and penetrations, and any necessary containment heat-removal systems shall be designed so that the containment structure can accommodate without exceeding the design leakage rate the pressures and temperatures resulting from the largest credible energy release following a loss-of-coolant accident, including a considerable margin for effects from metal–water or other chemical reactions that could occur as a consequence of emergency core-cooling systems.

50. *NDT Requirement for Containment Material.*[A] Principal load-carrying components of ferritic materials exposed to the external environment shall be selected so that their temperatures under normal operating and testing conditions are not less than 30° F above the nil ductility transition (NDT) temperature.

51. *Reactor Coolant-Pressure Boundary Outside Containment.*[A] If part of the reactor coolant-pressure boundary is outside the containment, appropriate features as necessary shall be provided to protect the health and safety of the public in case of an accidental rupture in that part. Determination of the appropriateness of features such as isolation valves and additional containment shall include consideration of the environmental and population conditions surrounding the site.

52. *Containment Heat-Removal Systems.*[A] Where active heat-removal systems are needed under accident conditions to prevent exceeding containment design pressure, at least two systems, preferably of different principles, each with full capacity, shall be provided.

53. *Containment Isolation Valves.*[A] Penetrations that require closure for the containment function shall be protected by redundant valving and associated apparatus.

54. *Containment Leakage-Rate Testing.*[A] Containment shall be designed so that an integrated leakage-rate testing can be conducted at design pressure after completion and installation of all penetrations and the leakage rate measured over a sufficient period of time to verify its conformance with required performance.

55. *Containment Periodic Leakage-Rate Testing.*[A] The containment shall be designed so that integrated leakage-rate testing can be done periodically at design pressure during plant lifetime.

56. *Provisions for Testing of Penetrations.*[A] Provisions shall be made for testing penetrations that have resilient seals or expansion bellows to permit leaktightness to be demonstrated at design pressure at any time.

57. *Provisions for Testing of Isolation Valves.*[A] Capability shall be provided for testing functional operability of valves and associated apparatus essential to the containment function for establishing that no failure has occurred and for determining that valve leakage does not exceed acceptable limits.

58. *Inspection of Containment Pressure-Reducing Systems.*[A] Design provisions shall be made to facilitate the periodic physical inspection of all important components of the containment pressure-reducing systems, such as pumps, valves, spray nozzles, torus, and sumps.

59. *Testing of Containment Pressure-Reducing-Systems Components.*[A] The containment pressure-reducing systems shall be designed so that active components, such as pumps and valves, can be tested periodically for operability and required functional performance.

60. *Testing of Containment Spray Systems.*[A] A capability shall be provided to test periodically the delivery capability of the containment spray system at a position as close to the spray nozzles as is practical.

61. *Testing of Operational Sequence of Containment Pressure-Reducing Systems.*[A] A capability shall be provided to test under conditions as close to the design as practical the full operational sequence that would bring the containment pressure-reducing systems into action, including the transfer to alternate power sources.

62. *Inspection of Air-Cleanup Systems.*[A] Design provisions shall be made to facilitate physical inspection of all critical parts of containment air-cleanup systems, such as ducts, filters, fans, and dampers.

63. *Testing of Air-Cleanup-Systems Components.*[A] Design provisions shall be made so that active components of the air-cleanup systems, such as fans and

dampers, can be tested periodically for operability and required functional performance.

64. *Testing of Air-Cleanup Systems.*[A] A capability shall be provided for in situ periodic testing and surveillance of the air-cleanup systems to ensure that (1) filter bypass paths have not developed and (2) filter and trapping materials have not deteriorated beyond acceptable limits.

65. *Testing of Operational Sequence of Air-Cleanup Systems.*[A] A capability shall be provided to test under conditions as close to design as practical the full operational sequence that would bring the air-cleanup systems into action, including the transfer to alternate power sources and the design air-flow delivery capability.

VIII Fuel- and Waste-Storage Systems

66. *Prevention of Fuel-Storage Criticality.*[B] Criticality in new and spent fuel storage shall be prevented by physical systems or processes. Such means as geometrically safe configurations shall be emphasized over procedural controls.

67. *Fuel- and Waste-Storage Decay Heat.*[B] Reliable decay-heat removal systems shall be designed to prevent damage to the fuel in storage facilities which could result in radioactivity release to plant operating areas or the public environs.

68. *Fuel- and Waste-Storage Radiation Shielding.*[B] Shielding for radiation protection shall be provided in the design of spent fuel- and waste-storage facilities as required to meet the requirements of 10 CFR 20.

69. *Protection Against Radioactivity Release From Spent Fuel and Waste Storage.*[B] Containment of fuel and waste storage shall be provided if accidents could lead to release of undue amounts of radioactivity to the public environs.

IX Plant Effluents

70. *Control of Releases of Radioactivity to the Environment.*[B] The facility design shall include those means necessary to maintain control over the plant radioactive effluents, whether gaseous, liquid, or solid. Appropriate holdup capacity shall be provided for retention of gaseous, liquid, or solid effluents, particularly where unfavorable environmental conditions can be expected to require operational limitations upon the release of radioactive effluents to the environment. In all cases, the design for radioactivity control shall be justified (1) on the basis of 10 CFR 20 requirements for normal operations and for any transient situation that might reasonably be anticipated to occur and (2) on the basis of 10 CFR 100 dosage-level guidelines for potential reactor accidents of exceedingly low probability of occurrence except that reduction of the recommended dosage levels may be required where high population densities or very large cities can be affected by the radioactive effluents.

Volatility Processes

By J. J. Barghusen

Nitrofluor Process

The Nitrofluor process for treating spent reactor fuels is based on the dissolution of the fuel in anhydrous hydrogen fluoride solutions containing complexing agents in the form of NO_2, N_2O_3, or NOF. These solutions are capable of dissolving a wide variety of fuels owing to the reactivity of the nitrosyl ion (NO^+) in the solvent. After the fuel is dissolved, the liquid phase is evaporated, and the solid residue is fluorinated to separate uranium and plutonium from fission products. All process steps are carried out in nickel or Monel equipment. An experimental study[1] was undertaken at Brookhaven National Laboratory (BNL) to determine the fundamental aspects of Monel and nickel corrosion in NO_2–HF solutions in the presence of air, water, and dissolved UO_2. Corrosion data were obtained by immersing nickel- and Monel-wire probes in the solutions and measuring the change in electrical resistance of the probe, which is related directly to change in the cross-sectional area of the wire.

In HF solutions containing $\leq$30 mole % NO_2, unstressed Monel and nickel were significantly attacked (total corrosion observed during test, $> 1 \times 10^{-3}$ mil) only at concentrations of 25 and 30 mole % NO_2. The rate and extent of corrosion were greater at 30 mole % NO_2. The mechanism of corrosion of both Monel and nickel in the system involves the growth of a superficial protective film that essentially precludes further corrosion. Although the initial rate of reaction of Monel and nickel in 30 mole % NO_2 solution at 100 to 150°C was high, the reaction rate decreased rapidly after 10 to 20 hr owing to formation of the protective film. The stability of the film was greater at the higher temperatures. In tests in which Monel was exposed to 30 mole % NO_2–70 mole % HF at 75°C, the surface film formed was soft and porous and was only partially protective; under these conditions the rate of corrosion did not decrease as the exposure increased.

The vapor-phase corrosion of both Monel and nickel was markedly increased by the presence of moisture. However, when the system was maintained above 100°C, thereby avoiding condensation, no significant corrosion was observed.

The presence of dissolved UO_2 or H_2O increased slightly the corrosion of Monel and nickel in the liquid phase. The extent of corrosion of Monel decreased with increasing temperature and amount of dissolved UO_2. The effect of stress on surface corrosion was insignificant in the elastic range, but in the plastic range it greatly increased both the initial corrosion rate and total corrosion during film formation.

On the basis of this study, it appears that both Monel and nickel are satisfactory materials for use in the Nitrofluor process. Nickel, in general, was superior to Monel under the experimental conditions owing to its lower corrosion rate and lower total corrosion prior to the formation of the protective film.

Studies on Irradiated High-Enriched Alloy Fuels

A fluid-bed fluoride-volatility process for the recovery of uranium from highly enriched uranium-alloy fuels (uranium–zirconium or uranium–aluminum alloy) is being developed at Argonne National Laboratory (ANL). The process involves two principal steps. In the first step the fuel is reacted with hydrogen chloride in a fluid-bed reactor to form volatile chlorides of zirconium ($ZrCl_4$) or aluminum ($AlCl_3$) which separate from nonvolatile uranium trichloride. In the next step, uranium is removed from the reactor as volatile UF_6 by fluorination with fluorine and is collected on sodium fluoride pellets. Further purification of the collected UF_6 can be achieved by desorption from NaF and collection of the UF_6 in cold traps. Chilenskas and associates[2] have summarized the results of process studies in a bench-scale fluid-bed facility using irradiated fuel materials. The experimental program was primarily directed toward obtaining information regarding (1) the distribution of fission products during the various process steps, (2) decontamination factors that may be realized, and (3) the effects, if any, of irradiation and the presence

of fission products on uranium recovery. Since complete details of the demonstration tests with irradiated alloy fuels reported in Ref. 2 were presented in an issue of *Reactor Fuel Processing*,[3] no further discussion of these experiments will be made here.

Fluid-Bed Fluorination of UO_2-PuO_2 Fuel with Chlorine Trifluoride

Of major concern in the development of fluoride-volatility processes for low-enrichment UO_2 fuel is establishing methods for the separation of uranium from plutonium and for the recovery of plutonium. Workers at Centre d'Étude de l'Énergie Nucléaire (CEN)[4] at Mol, Belgium, are developing a process for the recovery of uranium and plutonium from UO_2 fuel based on the selective fluorination of uranium to volatile UF_6 by the use of ClF_3 or ClF. Previous laboratory studies[5] showed that PuF_4 reacts with ClF_3 at 225 to 500°C to form volatile PuF_6 and that the reaction proceeds by the dissociation of ClF_3 at the solid surface producing ClF and fluorine atoms that, in turn, fluorinate PuF_4 to PuF_6. Since the dissociation of ClF_3 can be suppressed by the addition of chlorine to the system, a series of experiments was performed[4] to evaluate the use of ClF_3-Cl_2 mixtures for the separation of uranium from plutonium.

Several experiments were carried out to determine the reactivity of PuF_4 at 400°C toward ClF_3-Cl_2 mixtures of varying composition. It was observed that, for mixtures containing Cl_2/ClF_3 mole ratios less than 0.6, samples of PuF_4 exhibited a definite loss in weight, indicating volatilization of PuF_6. No such reaction was observed at 400°C between PuF_4 and mixtures in which the Cl_2/ClF_3 mole ratio was greater than 0.75, and at 550°C, with an equimolar mixture of ClF_3 and chlorine.

A single test was performed to determine the degree of separation achieved by contacting a mixture of UF_4 and PuF_4 with equimolar ClF_3-Cl_2 at 400°C. After 5 hr, over 96% of the uranium was converted to UF_6; no volatilization of plutonium was observed. At this point the flow of chlorine was stopped, and the PuF_4 residue was contacted with ClF_3 for 3 hr at 500°C. By means of this treatment, over 92% of the plutonium volatilized. This method appears promising for the separation of uranium and plutonium from a mixed tetrafluoride powder.

A series of laboratory-scale experiments was performed to determine the effect of temperature on the rate of fluorination of a solid mixture containing UF_4, UO_2F_2, and refractory alumina with a gas consisting of 5 vol.% ClF_3, 10 vol.% Cl_2, and 85 vol.% N_2. Within the temperature range 100 to 400°C, a definite maximum in the reaction rate was observed at 250°C.

The fluorination of solid mixtures of UF_4, UO_2F_2, and refractory alumina with gaseous mixtures of chlorine and ClF_3 has been studied in a 3.75-cm-diameter fluid-bed reactor. In tests in which 500 g of the solid mixture was contacted with a gas containing 10 vol.% Cl_2, 10 vol.% ClF_3, and 80 vol.% nitrogen, it was observed that the highest rate of fluorination occurred at 300°C. The fluorine utilization efficiency ranged from 61 to 82%. Slightly higher utilizations were effected by the use of a deeper bed. It was also observed that the reaction rate at 300°C was first order with respect to the concentration of ClF in nitrogen, and was essentially insensitive to the Cl_2/ClF_3 mole ratio at a constant ClF_3 flow rate.

The Use of Bromine Pentafluoride in the Processing of UO_2-PuO_2 Fuel

Workers at ANL are investigating a method for effecting the separation of uranium from plutonium based on the selective fluorination of uranium to UF_6 by bromine pentafluoride. The process involves three principal steps: (1) the oxide fuel, after a prior decladding step, is pulverized to $U_3O_8-PuO_2$ powder by reaction with oxygen at 450°C; (2) uranium is recovered from the oxide fines as gaseous UF_6 by reaction of the fines with ~10 vol.% BrF_5 at 300°C; and (3) plutonium, which is converted to solid PuF_4 in the previous step, is recovered as gaseous PuF_6 by reaction with fluorine at 300 to 550°C.

Jonke and associates[6] have discussed the application of the BrF_5 process to the reprocessing of low-enrichment fuels with specific emphasis on defining the current U. S. Atomic Energy Commission (AEC) program regarding volatility-process development work. The objective of the AEC program is to provide by 1970 the technical data required for the design of a commercial all-volatility plant having an initial capacity of 1 to 2 tons of fuel per day.

Summaries of work at ANL on the development of fluid-bed fluoride-volatility processes for low-enrichment oxide fuels have been recently published.[7,8] Since much of the information contained in these reports has been previously reviewed, only selected topics from these reports will be discussed.

Demonstration tests with UO_2-PuO_2 fuel on the process cycle $O_2-BrF_5-F_2$ are being carried out in a 2-in.-diameter fluid-bed reactor. A series of experiments has been completed to determine the effect of the BrF_5 fluorination temperature on plutonium recovery in the subsequent fluorination step. The results of initial experiments in this series were previously reviewed,[9] and the results indicated that the BrF_5 fluorination temperature does affect plutonium removal and that a BrF_5 fluorination temperature of 300°C is preferable to either 200°C or 400°C. Additional experiments[10,11] were performed at temperature levels of 250 and 350°C. In these experiments, 650 g of sintered $UO_2-0.5$ wt.% PuO_2 pellets, containing nonradioactive fission products, and 1100 g

of alumina, to which CsF, RbF, and NpO_2 were added, were charged to the fluid-bed reactor. The pellets were pulverized with 20 vol.% oxygen in nitrogen for 4 hr at 450°C. The oxide fines were reacted with 10 vol.% BrF_5 in nitrogen at 250 or 350°C for 2 hr. Plutonium fluorination with 90 vol.% fluorine was carried out for 3 hr at 300°C, for 5 hr while the temperature of the reactor was increased from 300 to 550°C, and for 3 hr at 550°C.

The course of uranium, plutonium, and neptunium volatilization during a typical experiment is shown in Fig. 1. In both experiments over 99.5% of the uranium in the charge volatilized during the BrF_5 fluorination step. After the plutonium fluorination step, the final alumina beds contained 0.008 wt.% plutonium for BrF_5 fluorination at 350°C and 0.007 wt.% for BrF_5 fluorination at 250°C. On the basis of these data and those reported previously,[9] it appears that, within the range 250 to 350°C, the removal of plutonium from the alumina bed is not affected by the temperature of the BrF_5 fluorination step. The results of these experiments also indicated that the addition of RbF and CsF to the system does not affect the removal from the alumina bed of uranium and plutonium by fluorination. As shown in Fig. 1, approximately 60% of the neptunium volatilized with the uranium during the BrF_5 fluorination step, and over 95% of the neptunium present in the charge was removed from the bed during both fluorination steps.

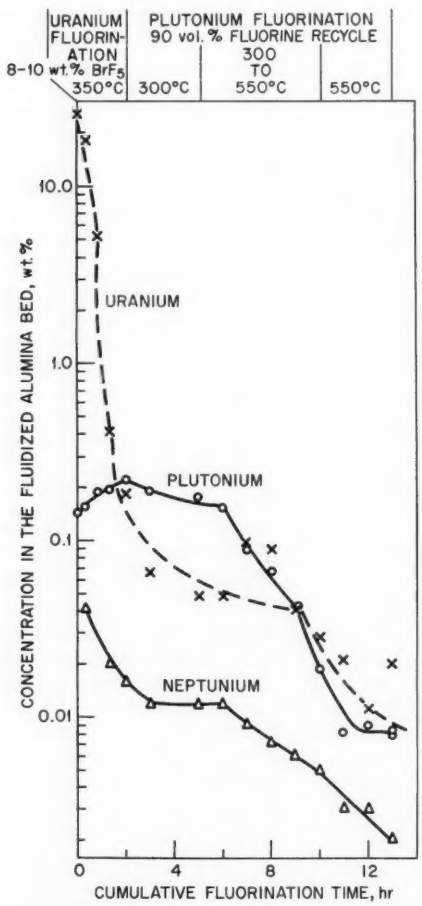
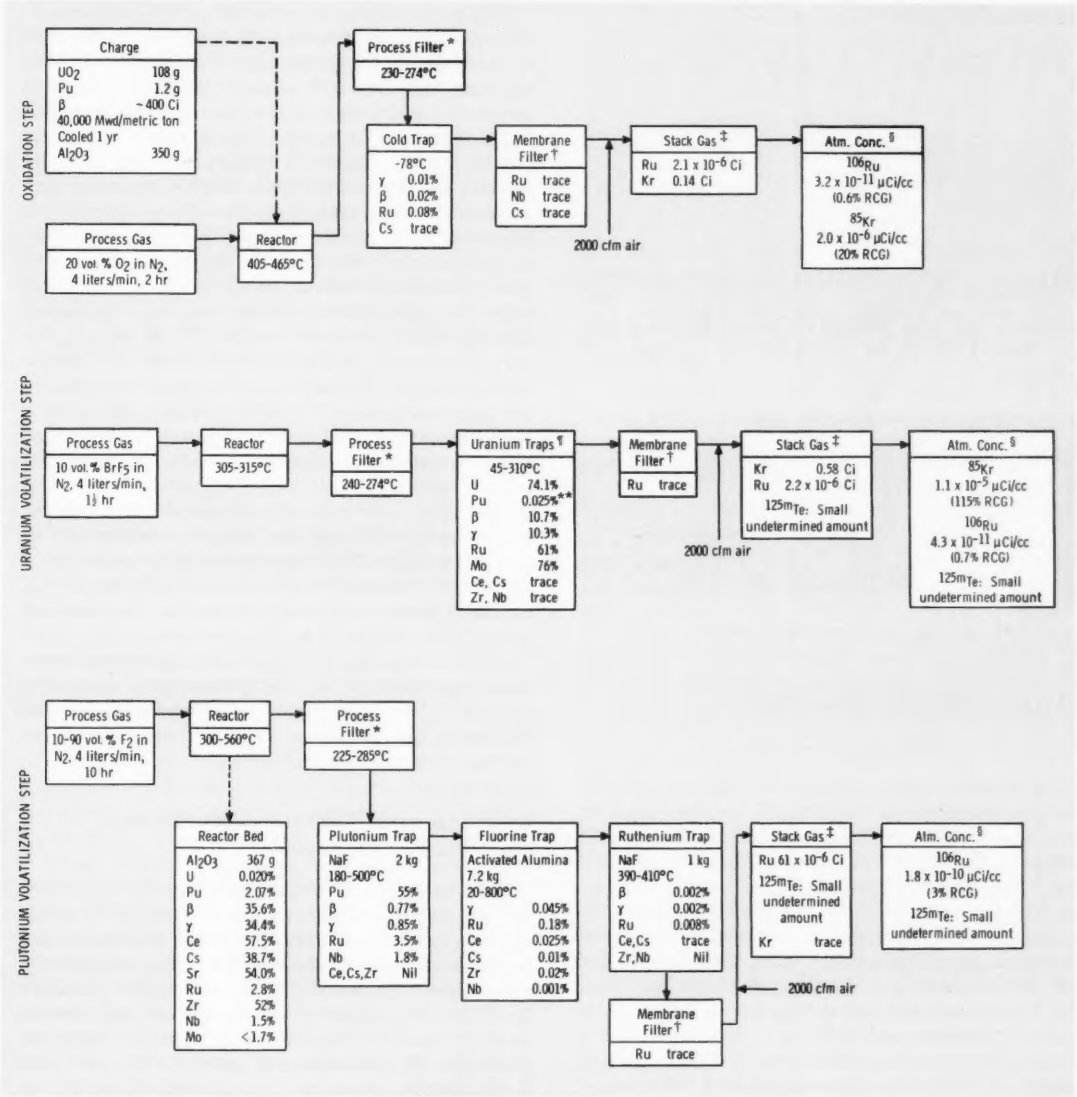


Fig. 1 Concentration of uranium, neptunium, and plutonium in alumina fluid bed during fluorination with BrF_5 followed by fluorine.[11]

A separate experiment was performed to evaluate the effect of high plutonium concentration in the oxide fuel on plutonium retention in the alumina bed.[12] In this experiment the oxide fuel pellets contained 5 wt.% plutonium, whereas previous experiments were performed with UO_2-PuO_2 pellets containing nominally 0.5 wt.% plutonium. The fuel charged for this experiment consisted of 651 g of 5 wt.% PuO_2-UO_2–fission-product pellets and 1100 g of 48- to 100-mesh alumina. The processing sequence was oxidation of the pellets for 4 hr at 450°C with 23 vol.% oxygen, fluorination of the uranium to UF_6 with 10 vol.% BrF_5 for 2 hr at 300°C, and recycle fluorination with 90 vol.% fluorine for 3 hr at 300°C, 3 hr at 350°C, 3 hr at 400°C, 2 hr at 450°C, 2 hr at 500°C, and 3 hr at 550°C.

The final alumina bed contained 0.004 wt.% uranium and 0.023 wt.% plutonium, and these amounts correspond to the removal of more than 99.9% of the uranium and 99% of the plutonium. Although the plutonium concentration in the final bed was greater than that obtained in previous experiments (0.007 wt.%) with oxide pellets containing 0.5 wt.% plutonium, the application of this fluoride-volatility process to oxide fuels containing up to 5 wt.% plutonium appears highly satisfactory since the percentage of plutonium removed from the bed in the case of the high-plutonium fuel was greater.

An experimental program is under way at ANL in a $1\frac{1}{2}$-in.-diameter fluid-bed reactor facility to determine the distribution of actinides and fission products in each of the process streams in the processing of irradiated oxide fuel by the $O_2-BrF_5-F_2$ process. A series of these experiments was completed[8] using a processing scheme consisting of oxidation of UO_2 to U_3O_8 at 400 to 450°C, uranium volatilization with BrF_5 at 300°C, and plutonium volatilization with fluorine at 300 to 560°C. The first of the experiments was performed with nonirradiated UO_2 to establish experimental procedures and to determine the levels of activity in the various samples which result from processing in equipment previously used for processing of radioactive fuels. The other experiments were made with declad UO_2 irradiated to 40,000 Mwd/metric ton and cooled for 1 year. The process conditions and the average fission-product distributions following each process step are shown in Fig. 2.



*Sintered nickel, 10-μ pore, grade G, Pall Trinity Micro Corporation.

†Membrane filter, 0.4-μ pore, Metricel VF-6 (fluorinated vinyl), Gelman Corporation.

‡The stack gas passed through on AEC high efficiency filter and a charcoal filter before it was sampled and discharged to the environment.

§The atmospheric concentrations at ground level were calculated from the stack gas analysis allowing a dilution factor of 10 from stack to ground level. The RCG value represents the percent of the recommended limiting concentration of the radioisotope as proposed in Radioactivity Concentration Guide (see Ref. 13).

¶The uranium traps consisted of two beds in series each containing ~1.3 kg of soda lime followed by a bed containing 0.85 kg activated alumina.

**Run BRF-1 only.

Fig. 2 Processing of irradiated UO_2 charges using an $O_2-BrF_5-F_2$ process scheme. Average values for two experiments.[8]

THE OXIDATION STEP

Only a small amount of activity volatilized during the conversion of the UO_2 to U_3O_8. The principal volatile activity was ^{85}Kr; about 17% of the total krypton activity was released during this step. It should be noted that the irradiated charges had been declad prior to processing and that the quantity of krypton released upon cladding removal is not known. Other volatile species detected after oxidation were trace amounts ($<1 \times 10^{-4}$% of that present in the charge) of cesium, niobium, and ruthenium. Analysis of the stack gas showed that, in addition to the krypton, ^{106}Ru was present at a concentration that was about 0.6% of the recommended limiting concentration.[13]

THE URANIUM-VOLATILIZATION STEP

During uranium volatilization by reaction with BrF_5, the principal activities that also volatilized and collected with the uranium in the soda-lime (a mixture of sodium hydroxide and hydrated lime) and activated-alumina traps were 61% of the ruthenium, 76% of the molybdenum, and trace amounts of Ce, Cs, Zr, and Nb. Also, the remainder of the krypton (83%) volatilized during the uranium fluorination step. In one experiment it was observed that 0.025% of the plutonium in the oxide fuel accompanied the uranium and collected in the soda-lime trap.[8]

THE PLUTONIUM-VOLATILIZATION STEP

The volatilization of plutonium from the reactor was accomplished by the use of fluorine at temperatures from 300 to 560°C. About 0.8% of the gross beta and gamma activity was volatilized during this step.[8] The major gamma active isotopes found with the plutonium in the NaF trap were ruthenium and niobium. Small amounts of cesium, cerium, and zirconium (less than 0.1%) were collected in the activated-alumina trap downstream of the NaF trap. The process off-gas contaminants were found to be mainly ruthenium and tellurium. About 3% of the recommended limiting value[13] for ^{106}Ru and a small amount of ^{125m}Te were found in the stack effluent.

REACTOR BED RESIDUES

The average values[8] (percentages of charged material) for the retention of actinides and fission products in the final alumina beds were 0.020% U, 2.0% Pu, 34.4% gross gamma, 35.6% gross beta, 57% Ce, 54% Sr, 39% Cs, 52% Zr, 1.5% Nb, and <1.7% Mo.

A comparison of the results for the inactive run and the two runs with irradiated charges indicated that the gamma activity of the samples obtained after processing with a nonirradiated charge is very small as compared with that obtained after processing an active charge, and therefore the contribution of activity from residual deposits within the equipment can be neglected.[8]

Implicit in the application of the BrF_5 process to the reprocessing of spent fuel materials is the need to consider methods for the recycle of the interhalogen. The effluent stream from the fluid-bed fluorinator will contain, in addition to UF_6 and excess BrF_5, elemental bromine, small amounts of volatile fission-product fluorides, and noncondensable gases such as oxygen and nitrogen. One method being considered for BrF_5 recycle involves (1) a regeneration vessel wherein bromine is converted to BrF_5 by fluorine at about 300°C, (2) a liquid condenser, and (3) a distillation system for the separation of BrF_5 from UF_6 and fission-product fluorides. Dunthorn[14] has prepared a detailed analysis of the design of a liquid condenser for this system. Since the liquidus temperature of the condensable gases in the condenser feed stream is about 0°C with a corresponding vapor pressure of 0.15 atm, it was necessary in this design analysis to consider dilution of the condensable gas stream with additional BrF_5. By adding 8 moles of diluent BrF_5 per mole of condensable gases in the condenser feed stream, the liquidus temperature is lowered to about −60°C and likewise the vapor pressure of the condensables is similarly reduced. The report[14] deals principally with the calculational procedures required in the design of the liquid condenser. Among the variables considered were the temperature of the inlet gas stream, the quantity of BrF_5 diluent, the quantity of condensables leaving the condenser as vapor, and the temperature and pressures within the condenser.

Environmental Contamination Control for Volatility Processing

Inherent in the development of volatility processes for reactor fuel materials is the need to provide reliable methods for the treatment of process gas streams containing fission-product fluorides and reagents such as interhalogens of fluorine. Workers at CEN[4] have investigated the use of soda lime to remove fluorine, chlorine, ClF_3, and ClF from gas streams. The experiments were carried out with 6- to 12-mesh soda lime at gas velocities of 2 and 6 cm/sec. In all cases the bed temperature at the start of the experiment was 220°C. It was observed that the capacity of soda lime for the four gases decreased with an increase in concentration and gas velocity. The optimum capacities per gram of soda lime were 0.3 g of F_2, 0.3 g of ClF_3, 0.23 g of ClF, and 0.53 g of Cl_2.

Preliminary experiments at CEN[4] have indicated that soda lime is also an effective agent for the removal of certain volatile fission-product fluorides from gas streams. Tellurium hexafluoride is readily fixed on soda lime at 200°C. Similarly, the volatile fluorides produced by fluorination of a mixture of MoO_3, ruthenium metal, and Sb_2O_3 with ClF_3 appear to be effectively sorbed on soda lime.

An experimental program is under way at ANL[8] to study the reactions between tellurium hexafluoride (TeF_6) and various solid materials. This study is directed toward determining the conditions whereby tellurium is fixed on the solid sorbent. In a survey of various sorbent materials, the effectiveness of the solid for removal of TeF_6 from a process gas stream was evaluated in terms of the rate of TeF_6 sorption, the capacity of the solid for TeF_6, and the retention of TeF_6 on the solid under vacuum. The results of this survey indicated that activated alumina, Linde 13X Molecular Sieve, BPL activated charcoal, magnesium fluoride, and copper-metal turnings are promising reagents for the removal of TeF_6 from gas streams.

In recent studies of the removal of TeF_6 from process gas streams using activated alumina as the solid sorbent, it was found that the required tellurium decontamination factor of 700 can be readily achieved under a variety of conditions.[15] In these studies several process conditions were varied: temperature of the activated alumina, 25 and 100°C; bed height, 1 and 2 in.; velocity of gas through the alumina bed, 20 and 40 ft/min; and concentration of TeF_6 in the gas stream, 250 and 500 ppm. The results indicated that only the bed height had a significant effect on the tellurium decontamination factor. Gas flow rate and TeF_6 concentration showed only a slight effect on tellurium removal.

In connection with work at ANL on the chemistry of tellurium fluorides, a literature survey of the properties of the oxygen and fluorine compounds of tellurium was prepared and published in the form of a report.[16]

Other work at ANL has concerned an investigation of the factors that affect the filtration of particulate material by high-efficiency filters. This work has been specifically directed toward analyzing the filtration behavior of the aerosol generated by the reaction between PuF_6 and atmospheric moisture to obtain information for the design of efficient containment and gas cleanup systems. The hydrolysis of UF_6 and PuF_6 proceeds through the formation of solid UO_2F_2 and PuO_2F_2, respectively. The kinetics of these reactions have been measured by Kessie;[17] results of these studies were briefly reviewed in the Winter 1965–1966 issue of *Reactor Fuel Processing*.[18]

Plutonium hexafluoride, when released to the atmosphere, reacts with moisture in the air to form fine particles (<0.1 μ in diameter) of PuO_2F_2. Up to 60% of this plutonium will remain airborne. The combination of high dispersibility and low permissible concentration (3×10^{-8} mg of ^{239}Pu per cubic meter of air) requires a reduction in plutonium concentration by filtration or other processes by a factor of less than 10^{-6}. Because of the lack of information on the effectiveness of filters in removing particulate material from gas streams in situations requiring very low penetrations, an intensive study was conducted at ANL to evaluate the performance of filters with PuF_6–air mixtures. The results of this study have been reported by Kessie.[19] It was observed in this work that, with air containing excess moisture, the fraction of released plutonium penetrating two high-efficiency filters in series amounts to between 10^{-9} and 10^{-6} at a gas velocity of 1 cm/sec. Filter efficiency was found to increase by the agglomeration of particles before filtration and by loading effects of collected particles on the filters. When an excess of moisture was present, agglomeration effects tended to limit the maximum discharge rate of plutonium, and loading effects limited the maximum total quantity of plutonium that could be discharged from a single filter.

As part of the study of the containment of PuF_6 within ventilated enclosures, a computer program has been developed to obtain numerical solutions of differential equations describing the mechanisms occurring in filtration.[20] In this program the fiber efficiency (directly related to the filter penetration) was calculated as a function of several variables: fiber properties—diameter, spacing, charge, dielectric constant; gas properties—viscosity, density, mean free path, velocity, dielectric constant; particle properties—diameter, density, temperature, charge, dielectric constant. A series of computations has been made to determine fiber efficiency for average conditions present in typical high-efficiency filters: fiber diameter (D_F), 0.3 μ (geometric mean); fiber spacing (S_F), 1.8 μ; fiber surface area per unit area of filter media, 77. Typical results at two levels of gas velocity (V_G), 2.5 cm/sec (100% rated flow) and 0.25 cm/sec (10% rated flow), are shown in Fig. 3. The two fiber geometries: $F_Y = 0$ for a rectangular array and $F_Y = \frac{1}{2}$ for a triangular array show very different efficiencies particularly at the higher gas velocities. It is significant that the particle diameter for minimum efficiency or maximum penetration in all cases was some diameter greater than 0.2 μ. The experimentally measured di(2-ethylhexyl)phthalate (DOP) penetrations of 1×10^{-5} to 5×10^{-4} for 0.3-μ-diameter DOP particles at V_G of 2.5 cm/sec are between the two curves for $F_Y = 0$ and $F_Y = \frac{1}{2}$. The calculated results are essentially the same for particles with densities of 1 and 6.5 g/ml, which represent the densities of DOP and plutonyl fluoride, respectively.

Boback and Heatherton[21] have described the bioassay aspects of a UF_6 release, and their findings are pertinent to the problem of environmental contamination as related to fluoride-volatility processing. The release occurred when a valve was accidentally removed from a heated 10-ton UF_6 cylinder. Approximately 3800 lb of uranium, as UF_6, escaped before the opening was plugged. Most of the UF_6 which escaped was absorbed in the spray from fire hoses which was directed on the escaping gas. Although several persons were exposed to the escaping gas

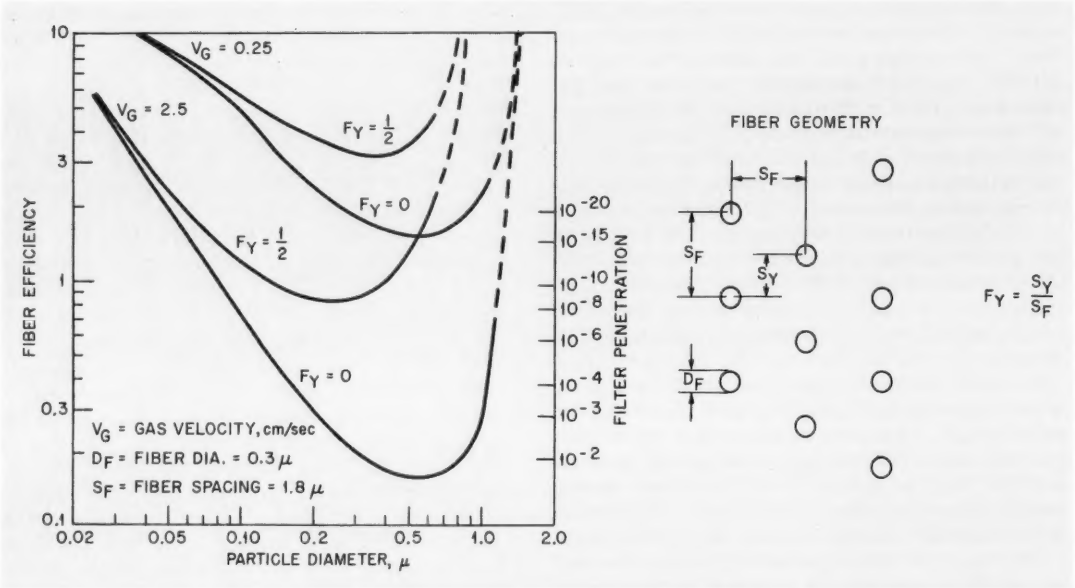


Fig. 3 Results of fiber efficiency computation.[20]

and UO_2F_2 fume, no clinical abnormalities were found. In all cases the uranium concentration in the urine returned to normal levels within 80 hr. One case of pulmonary edema developed; however, this individual was fully recovered within 9 days.

Corrosion

Owing to the high temperatures and the highly reactive nature of the principal reagents used in fluoride-volatility processes, corrosion of equipment components must be carefully considered in the design of processing facilities. Workers at CEN[4] have evaluated the corrosion resistance of several metals and alloys in ClF_3 at 200°C. All specimens were exposed to static ClF_3 for 1920 hr, and the rate of penetration was determined by weight change of the specimen. Aluminum, Corronel 280, copper, Incoloy 825, Inconel, Monel, and nickel showed good resistance (< 5 μ/year penetration) to ClF_3 at 200°C. Soft steel, Hastelloy B, Hastelloy C, and stainless steels 302 and 304 exhibited higher rates of corrosion.

Gunther and Steindler[22] have summarized the results of an investigation to determine the rates of corrosion of nickel and nickel alloys at 500°C by UF_6 and several volatile fission-product fluorides either individually or in the presence of fluorine. In general, it was observed that Nickel 200 and Monel exhibited the best overall resistance to corrosion by fluorine, UF_6, and fission-product fluorides.

In a detailed study of the reaction between fluorine and nickel and Monel, Riwan and Auguin[23] analyzed the structures of the fluoride surface films. On the basis of these analyses, the authors suggest that the reaction proceeds by the simultaneous diffusion of fluoride and nickel ions.

Reactions of Fluorinating Agents with Water

Workers at the Paducah Gaseous Diffusion Plant[24,25] have investigated the reaction between ClF_3 and H_2O. Results of previous studies[26] showed that the initial reaction between ClF_3 and H_2O proceeds according to the equation

$$2ClF_3 + 2H_2O \rightarrow 4HF + ClF + ClO_2F$$

Since both ClF and ClO_2F react with H_2O, studies were continued to investigate the nature of these reactions. In the temperature range 60 to 160°C, the products[24] of the vapor-phase reaction between ClF and H_2O are HF, O_2, Cl_2, and Cl_2O in the presence of excess H_2O. With excess ClF present, ClO_2F is formed rather than Cl_2O. It is postulated that the initial reaction is $2ClF + H_2O \rightarrow Cl_2O + 2HF$. In the presence of excess ClF, further reaction occurs as $2Cl_2O + ClF \rightarrow ClO_2F + 2Cl_2$. This system is complicated by other reactions such as the decomposition of Cl_2O and ClO_2F and the reaction of ClO_2F with H_2O.

It has been observed[25] that the vapor-phase reaction of ClO_2F with water at 63 and 155°C proceeds by the formation of ClO_2 as indicated by the equation $4ClO_2F + 2H_2O \rightarrow 4HF + 4ClO_2 + O_2$. Since ClO_2 thermally decomposes and produces Cl_2, Cl_2O, and ClO_3,

the complete analysis of the system is complicated. At 155°C no ClO_2 was observed; the only volatile products were O_2, Cl_2, and HF. Thus the overall reaction after decomposition of intermediate compounds may be best represented by the equation

$$4ClO_2F + 2H_2O \rightarrow 4HF + 2Cl_2 + 5O_2$$

References

1. A. Kigoshi and G. Strickland, Monel and Nickel Corrosion in Nonaqueous NO_2–HF Solutions, USAEC Report BNL-9409, Brookhaven National Laboratory, August 1966.
2. A. A. Chilenskas, K. S. Turner, J. E. Kincinas, and G. L. Potts, Engineering Development of Fluid-Bed Fluoride-Volatility Processes. Part 10. Bench-Scale Studies on Irradiated, Highly Enriched, Uranium-Alloy Fuels, USAEC Report ANL-6994, Argonne National Laboratory, October 1966.
3. *Reactor Fuel Process.*, 9(4): 202-205 (Fall 1966).
4. Centre d'Étude de l'Énergie Nucléaire, Reprocessing of Irradiated Fuels, Quarterly Report No. 25, April 1–June 30, 1966, USAEC Report EURAEC-1785, 1967.
5. *Power Reactor Technol. Reactor Fuel Process.*, 10(1): 72 (1967).
6. A. A. Jonke, M. Levenson, N. M. Levitz, M. J. Steindler, and R. C. Vogel, Fluoride Volatility, Candidate for Second-Generation Fuel-Reprocessing Plants, *Nucleonics*, 25(5): 58-63 (May 1967).
7. Argonne National Laboratory, Chemical Engineering Division Semiannual Report January–June 1966, USAEC Report ANL-7225, November 1966.
8. Argonne National Laboratory, Chemical Engineering Division Research Highlights, May 1966–April 1967, USAEC Report ANL-7350, 1967.
9. *Power Reactor Technol. Reactor Fuel Process.*, 10(1): 75 (1967).
10. R. M. Adams and A. Glassner (Comps.), Reactor Development Program Progress Report, December 1966, USAEC Report ANL-7286, p. 51, Argonne National Laboratory, Jan. 26, 1967.
11. R. M. Adams and A. Glassner (Comps.), Reactor Development Program Progress Report, February 1967, USAEC Report ANL-7308, pp. 71-73, Argonne National Laboratory, Mar. 28, 1967.
12. R. M. Adams and A. Glassner (Comps.), Reactor Development Program Progress Report, March 1967, USAEC Report ANL-7317, pp. 83-84, Argonne National Laboratory, Apr. 28, 1967.
13. *Radioactivity Concentration Guide*, AEC Manual, Chap. 0524, Standards for Radiation Protection (approved Aug. 12, 1963). Copy can be obtained from AEC Headquarters, Division of Operational Safety, Washington, D. C.
14. D. I. Dunthorn, Liquid Condensation of a Uranium Hexafluoride–Bromine Pentafluoride Mixture from a Noncondensable Carrier Gas Stream, USAEC Report K-L-2223, Oak Ridge Gaseous Diffusion Plant, Feb. 1, 1967.
15. R. M. Adams and A. Glassner (Comps.), Reactor Development Program Progress Report, April 1967, USAEC Report ANL-7329, p. 99, Argonne National Laboratory, June 16, 1967.
16. D. R. Vissers and M. J. Steindler, Laboratory Investigations in Support of Fluid-Bed Fluoride-Volatility Processes. Part X. A Literature Survey on the Properties of Tellurium, Its Oxygen and Fluorine Compounds, USAEC Report ANL-7142, Argonne National Laboratory, February 1966.
17. R. W. Kessie, Plutonium and Uranium Hexafluoride Hydrolysis Kinetics, *Ind. Eng. Chem., Process Design Develop.*, 6(1): 105-111 (January 1967).
18. *Reactor Fuel Process.*, 9(1): 29-31 (Winter 1965–1966).
19. R. W. Kessie, Removal of Plutonium Hexafluoride from Cell Exhaust Air by Hydrolysis and Filtration, in Proceedings of the Ninth AEC Air Cleaning Conference held in Boston, Massachusetts, September 13–16, 1966, J. M. Morgan, Jr., and M. W. First (Eds.), USAEC Report CONF-660904, Vol. 2, pp. 741-762, January 1967.
20. R. M. Adams and A. Glassner (Comps.), Reactor Development Program Progress Report, February 1967, USAEC Report ANL-7308, pp. 93-95, Argonne National Laboratory, Mar. 28, 1967.
21. M. W. Boback and R. C. Heatherton, Bioassay Aspects of a UF_6 Fume Release, USAEC Report NLCO-986, National Lead Company of Ohio, 1966.
22. W. H. Gunther and M. J. Steindler, Laboratory Investigations in Support of Fluid-Bed Fluoride-Volatility Processes. Part XIV. The Corrosion of Nickel and Nickel Alloys by Fluorine, Uranium Hexafluoride, and Selected Volatile Fission-Product Fluorides at 500°C, USAEC Report ANL-7241, Argonne National Laboratory, December 1966.
23. R. Riwan and B. Auguin, Fluoride Film Structures on Monel and Nickel, *Compt. Rend. Ser. C. Chim Sci.*, 264: 725-728 (Feb. 27, 1967).
24. W. R. Rossmassler (Ed.), Paducah Plant Laboratory Progress Report January–June 1966, USAEC Report KY-L-419 (Pt. 1), Paducah Gaseous Diffusion Plant, Sept. 27, 1966.
25. W. R. Rossmassler (Ed.), Paducah Plant Laboratory Progress Report July–December 1966, USAEC Report KY-L-419 (Pt. 2), Paducah Gaseous Diffusion Plant, Apr. 17, 1967.
26. *Reactor Fuel Process.*, 9(3): 161 (Summer 1966).

Compact Pyrochemical Processes

By T. R. Johnson

Fuel-recovery processes in which the separations of the fuel materials from the fission products are made in liquid metal–liquid salt media are classified generally as pyrochemical processes because the usual operating temperatures are in the 500 to 800°C range. Pyrochemical processes are capable of treating short-cooled, high-burnup fuels in compact process equipment. Most of the development work on pyrochemical processes is directed toward the recovery of fast breeder reactor fuels because the advantages of these processes should be particularly beneficial to the recovery of such fuels.

Liquid Metal–Salt Extraction Processes

In several pyrochemical processes currently being developed, the separation of fertile and fissile elements from the fission products depends on equilibrium oxidation–reduction reactions between a liquid metal and a liquid salt. The oxidized species are dissolved or suspended in the liquid-salt phase, whereas the reduced species are in the metal phase. Separations can be made because of differences in the distribution of the various elements between the liquid-metal and liquid-salt phases. Typical liquid-metal solvents are alloys of Mg with Cu, Cd, or Zn; typical salts are mixtures of the chlorides and fluorides of alkali metals and alkaline earths. In many of these extraction processes, magnesium is the reducing agent, and magnesium chloride is the oxidant.

SALT-TRANSPORT PROCESSES

Current investigations of pyrochemical processes at Argonne National Laboratory (ANL) are related to flow sheets that incorporate a salt-transport step. The salt-transport step involves the selective extraction of solutes from one solvent metal (the donor) into a second metal (the acceptor) through a molten salt (the carrier salt). If the proper compositions of the donor and acceptor alloys and the carrier salt are employed, uranium and plutonium can be separated from zirconium and the more noble fission products, and plutonium can be separated from uranium. In the flow sheet presently under development at ANL (described in an earlier issue of *Power Reactor Technology and Reactor Fuel Processing*[1]), the donor alloy is a Cu–33 wt.% Mg alloy, the carrier salt is a mixture containing $MgCl_2$, and the acceptor alloy is a Zn–5 wt.% Mg alloy. In the present flow sheet, the alkali, alkaline earth, and lanthanide elements are separated from the actinides in the donor alloy by multistage, liquid metal–liquid salt extraction prior to the salt-transport step. During the salt-transport step, uranium and the noble-metal fission products remain in the donor alloy, and plutonium and any residual electropositive fission products are extracted into the acceptor alloy. The uranium, which is insoluble in the Cu–Mg alloy, is isolated from salt, copper, and fission products by decantation and then by washing with liquid magnesium.

A laboratory-scale salt-transport experiment was performed[2] using apparatus that minimized the cross contamination between the donor alloy and the acceptor alloy. In this apparatus the molten-salt and liquid donor alloy were stirred together and allowed to settle. Then the furnace was tilted, and the salt was top poured into an adjacent crucible that contained the acceptor alloy. After the salt and the acceptor alloy were stirred and allowed to separate, the salt was top poured back into the crucible containing the donor alloy. In the experiment, cerium, which was used as a stand-in for plutonium, was transferred from a Cu–33 wt.% Mg donor alloy through a 50 mole % $MgCl_2$–30 mole % NaCl–20 mole % KCl carrier salt to a Zn–5 wt.% Mg acceptor alloy at 600°C. After five cycles, 93% of the cerium charged to the Cu–Mg alloy had been transferred to the Zn–Mg alloy. It was estimated that, with an additional five cycles, 99.5% of the cerium would have been transferred. The copper content of the Zn–Mg alloy remained constant at about 0.088 wt.% during the last three cycles; therefore copper transfer to the acceptor alloy was small.

The distribution of cerium between liquid Zn–Mg alloys and liquid 50 mole % $MgCl_2$–30 mole % NaCl–20 mole % KCl was measured[2] at 600, 700, and 800°C over a range of magnesium concentrations in the metal phase. The distribution coefficients shown in Fig. 1 indicate that, if Zn–Mg alloys are used as acceptor alloys, any cerium, and presumably other rare earths, not removed before the salt-transport step will follow the plutonium into the Zn–Mg alloy.

The first process step after the decladding of a mixed oxide (PuO_2–UO_2) fuel is the reduction of the oxides to metal by a Cu–33 wt.% Mg alloy in contact with a molten salt. After reduction the actinides and the noble and refractory-metal fission products are in the metal phase. The plutonium is in solution, and the uranium is a precipitate. Experiments were performed to measure the solubility of uranium and fission products in the Cu–Mg alloy and to determine if fission products would coprecipitate with uranium. The solubilities determined[3] in liquid Cu–33 wt.% Mg were as follows: U, 0.023 wt.% (600°C) and 0.22 wt.% (800°C); Mo, ~0.001 wt.% (600 to 850°C); Nb, <0.002 wt.% (600 to 850°C); Ru, 0.6 wt.% (600°C) and 1.5 wt.% (850°C). It was also determined[2] that zirconium does not coprecipitate significantly with beta or gamma uranium but that it may do so with alpha uranium.

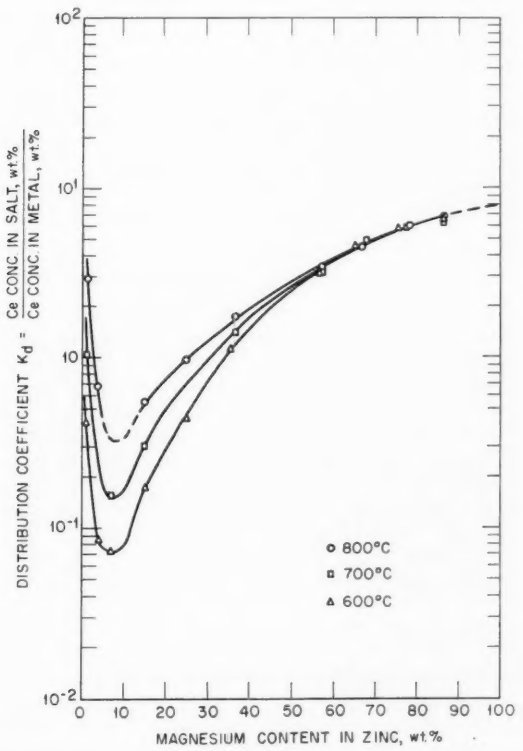


Fig. 1 Distribution[2] of cerium between 50 mole % $MgCl_2$–30 mole % NaCl–20 mole % KCl salt and Zn–Mg alloys at 600, 700, and 800°C.

Because spent reactor fuel will contain neptunium, experiments were made to determine the behavior of this element in the reduction step of the salt-transport process. Experiments[4] showed that NpO_2 was reduced by the Cu–33 wt.% Mg alloy and that, when NpO_2 and UO_2 were coreduced, some neptunium coprecipitated with the uranium. The distribution of neptunium between a 50 mole % $MgCl_2$–30 mole % NaCl–20 mole % KCl molten-salt mixture and a liquid Zn–10 wt.% Mg alloy was also measured. The distribution coefficient (wt.% neptunium in salt/wt.% neptunium in metal) varied from 2.3×10^{-3} at 800°C to 7.8×10^{-3} at 574°C. Because the distribution coefficients of neptunium and plutonium diverge below 700°C, it may be possible to separate these elements by liquid–liquid extraction. In the same experiment, neptunium solubility in the Zn–10 wt.% Mg alloy was found to be 8.1 wt.% at 800°C and 0.2 wt.% at 574°C.

ALTERNATIVE PROCESSES FOR MOLTEN SALT BREEDER REACTOR FUEL

The latest progress report[5] of the Molten Salt Breeder Reactor (MSBR) program summarizes investigations of possible pyrochemical methods of reprocessing fluoride fuels from these reactors. The investigations include (1) the reduction of rare earths from the salt into a lead or bismuth alloy, (2) the removal of rare earths from the salt by precipitation with UF_3, and (3) the removal of protactinium from blanket melts by precipitation with ZrO_2 or by reduction with thorium. Blankenship[6] has also discussed this work from the point of view of maximizing the breeding gain of a thermal breeder.

A liquid–liquid extraction process is being investigated as a means of removing the rare-earth fission products from the fuel solvent of the reference-design Molten Salt Breeder Reactor. This process would follow removal of the uranium from the salt by fluoride-volatility techniques. Essentially all the Ce, La, and Nd and substantial quantities of Sm and Eu can be removed from liquid Li_2BeF_4 by a Bi–2 mole % Li alloy. In the conceptual process these fission products would be back extracted and concentrated in another salt mixture by hydrofluorination. Because some isotopically pure ^7Li is lost in this process, the objective of recent experiments has been the conservation of the lithium reductant in the metal–salt extraction step. The stability of Li–Bi alloys at 600°C in low-carbon steel was first demonstrated. When purified Li_2BeF_4 was added to stable Li–Bi alloys, a loss of lithium from the metal phase occurred that corresponded to 0.11 mole of lithium per kilogram of salt. This loss should not materially affect the economics of the rare-earth extraction process.

When rare earths were extracted from the liquid Li_2BeF_4 into bismuth by the addition of beryllium metal, there was also an increase in the lithium concentration of the metal phase. Further studies of the equilibrium between LiF and beryllium in the two-phase system were begun.

Protactinium is removed from solution in a mixture of Li_2BeF_4 and ZrF_4 by the addition of ZrO_2. Additional experiments were made to elucidate the mechanism of this reaction. In each experiment, incremental amounts of ZrO_2 particles of specific surface area were added at 600°C to a salt that contained ^{233}Pa. It was determined that protactinium removal from the salt mixture was not proportional to the surface area of the added oxide and that the removal did not conform to an equilibrium solid solution of protactinium in ZrO_2.

Protactinium has also been removed from a molten mixture of $LiF-2$ mole % BeF_2-25 mole % ThF_4 by adding thorium either as the metal or as a solution in lead or bismuth. The reduced protactinium species did not form a stable liquid-metal solution but was deposited on vessel walls that were in contact with the salt. An experimental pump loop (Fig. 2) was designed to contact a fluoride mixture containing dissolved ^{233}Pa with a stream of molten bismuth containing thorium. The thorium was introduced by passing the bismuth over thorium chips and then spraying the alloy into the top of the vessel containing the salt. The protactinium extracted at the surfaces of the free-falling bismuth droplets was removed by pumping the metal through a filter formed by fine steel wool. During operation of the loop, 96% of the ^{233}Pa originally in the salt phase was removed by adding thorium metal, and at least 43% was collected on rather small volumes of steel wool. However, about 50% of the protactinium could not be accounted for, and, although large amounts of thorium were added to the system, its concentration in the bismuth remained very low. Other experiments showed that macroquantities of ^{231}Pa dissolved in a fluoride salt were readily reduced by thorium but that a large fraction of the reduced protactinium remained suspended in the molten $LiF-ThF_4$ mixture.

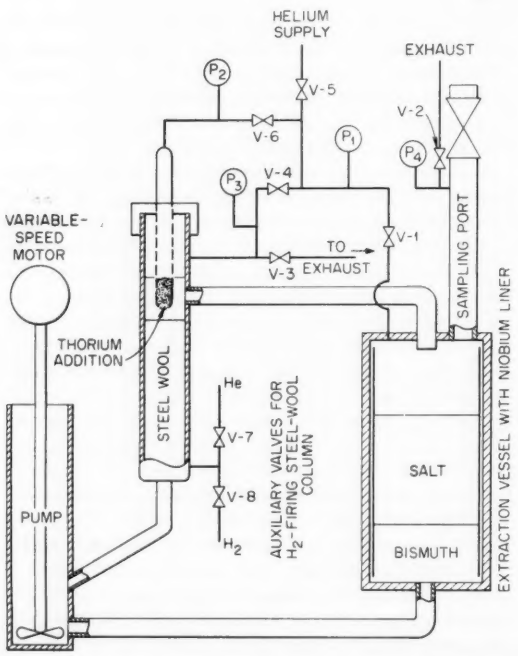


Fig. 2 Diagram of ^{233}Pa extraction pump loop.[5]

SLAGGING PROCESSES

High-temperature slagging processes involve the selective oxidation of more reactive components from a liquid-metal phase that is usually molten uranium or plutonium. Melt refining, which is an example of an oxide slagging process, is used routinely to reprocess fuel discharged from EBR-II. The production performance of this melt-refining operation is summarized in monthly progress reports of ANL.[2-4]

Halide slagging processes involve the reaction of metal phase components with components of a liquid chloride or fluoride salt. A slagging procedure[7] employing UCl_3 as an oxidant was studied as an attractive means of extracting plutonium from molten U–Pu alloys. The salt containing the plutonium and the more electropositive elements would be a suitable feed for decontamination and recovery by conventional or pyrochemical processing methods. The equilibrium of the reaction

$$UCl_3 + Pu \rightleftharpoons PuCl_3 + U$$

was determined by contacting molten uranium containing 0.2 to 4.2 mole % plutonium with $CaCl_2$ containing UCl_3. Equilibrium constants, based on mole and atom fractions, were about 200 at 1150°C and 170 at 1200°C. At 1200°C the loss of plutonium by volatilization was about 1% in 30 min, and the contamination of the metal ingot with beryllium from the BeO containment crucible was negligible. The equilibrium constant for the reaction at 1200°C was sufficiently large that plutonium bred in metallic blanket fuel could be extracted by halide slagging.

MISCELLANEOUS DEVELOPMENTS

The selection of suitable materials for processing equipment is an important consideration in pyrochemical processes that employ zinc-rich solvent alloys and molten chloride salts at temperatures as high as 800°C. One such process is skull reclamation,[8] which was studied at ANL as a means of recovering uranium metal from a mixture of oxides of uranium and fis-

sion products. For development work on this process, tungsten was used as a crucible material, and a 70 wt.% Mo–30 wt.% W alloy was used for applications requiring a more malleable material. Tungsten has been shown[9] to possess excellent corrosion resistance to zinc–salt systems, and the Mo–W alloy has been found to possess adequate corrosion resistance for use as agitators and process-solution transfer lines. Several methods of fabrication were employed to produce crucibles that were tested under process conditions. Tungsten crucibles made by isostatic pressing and sintering or by shear forming gave excellent service and reliability. Crucibles formed by plasma-spraying a tungsten powder on a mandrel showed promise, but crucibles formed by spraying tungsten powder on a substrate or by arc-welding tungsten sheet were found to be unsatisfactory.

Developments related generally to metal–salt extraction processes have appeared recently in the literature. A German patent[10] was issued for a method of separating fission products from an oxide fuel. The contaminated uranium oxide is mixed with a series of volatile metal–salt mixtures that progresses from weak to strong reductants. The metal alloys are then evaporated under vacuum to recover the reduced fission products.

A procedure for recovering waste salt from a pyrochemical process was disclosed in a U. S. patent.[11] In this procedure fission products, especially the rare earths, are extracted from a molten-halide-salt phase into a binary alloy of aluminum and magnesium. The salt and metal phases are separated. The purified salt can then be reused, and the metal phase provides convenient and compact containment of the fission products.

Electrolytic Processes

SALT-CYCLE PROCESS

The salt-cycle process was developed at Hanford to reprocess short-cooled UO_2–PuO_2 fuels for recycle to thermal reactors. The principal step in this process is the electrolytic deposition of crystalline, reactor-grade UO_2–PuO_2 solid solutions from molten chloride salts. Equipment for remote operation of the process was designed and tested, and several hundred pounds of UO_2 that met reactor specifications was produced electrolytically for evaluation as nuclear fuel. Two terminal reports from Battelle–Northwest summarize the engineering development of the various operations of the salt-cycle process[12] and describe a process demonstration with irradiated UO_2–PuO_2 fuel.[13] Earlier developments of the salt-cycle process were presented in issues of *Reactor Fuel Processing*.[14]

One version[13] of the salt-cycle process that is applicable to a close-coupled plutonium recycle system is shown in Fig. 3. In addition to the codeposition of UO_2–PuO_2, the process provides for an alternative step in which partitioning of uranium and plutonium is achieved by electrodeposition of UO_2. The engineering development[12] involved pilot-plant productions of electrodeposited UO_2 and the design and evaluation of equipment for remote recovery of irradiated mixed-oxide fuel.

Vibratory decladding with an air hammer was selected[12] as the primary technique for fuel-cladding separation because of its effectiveness in removing all the fuel from the cladding, and the minimal waste-disposal problems. For cold-swaged UO_2, decladding times as short as 2 min for a 0.5-in.-diameter by 4-ft-long rod were demonstrated in remotely operable apparatus with no appreciable formation of dust. Scouting studies of oxidative decladding and of decladding of Zircaloy-2 and stainless steel by chlorine gas or lead chloride in molten salts were also made.

Uranium dioxide was oxidized to U_3O_8 in remotely operable equipment to promote dissolution in the salt bath by reducing the particle size of the oxide. For 15-lb batches of UO_2, a flow rate of 2 liters of air per minute for 24 hr was sufficient to prepare feed for dissolution, although only partial oxidation to U_3O_8 was accomplished throughout the oxide bed in this time. Sintering of the oxide during temperature excursions was a major problem.

A LiCl–KCl system was selected instead of a NaCl–KCl or a $PbCl_2$–KCl system as being most suitable for the oxide-dissolution and -deposition steps. The use of LiCl–KCl permitted operating temperatures of 500 to 600°C, a range that minimized corrosion by chlorine in the vapor space. High oxide-dissolution rates were achieved in this salt by chlorine sparging with a submerged gas lift. With improved atmosphere control the deposited oxides had oxygen-to-uranium ratios below 2.01, high densities, good crystal structure, and chloride contents in the range 50 to 100 ppm.

Cathodes that were made of graphite with an impervious pyrolytic graphite coating were used and were found to facilitate removal of the deposited oxides and to give a product that contained less than 50 ppm carbon. Metal, graphite, and ceramic materials were evaluated for use as containers. All metals corroded excessively, and graphite prevented Pu(IV) from oxidizing to Pu(VI) in the codeposition step. Fused silica was chosen as the container material.

From studies of the codeposition and partition steps using cerium as a stand-in for plutonium, it was concluded that the O–Cl sparges necessary to oxidize plutonium during electrolysis were compatible with the production of UO_2 deposits that had good crystal structures and proper oxygen contents.

Integrated crushing, grinding, and sieving devices that produced three narrow particle-size distributions required for vibratory compaction were evaluated for hot-cell use. Chlorine and hydrogen chloride were removed from the off-gas by caustic scrubbers in all pilot-plant operations. The recovery of chlorine

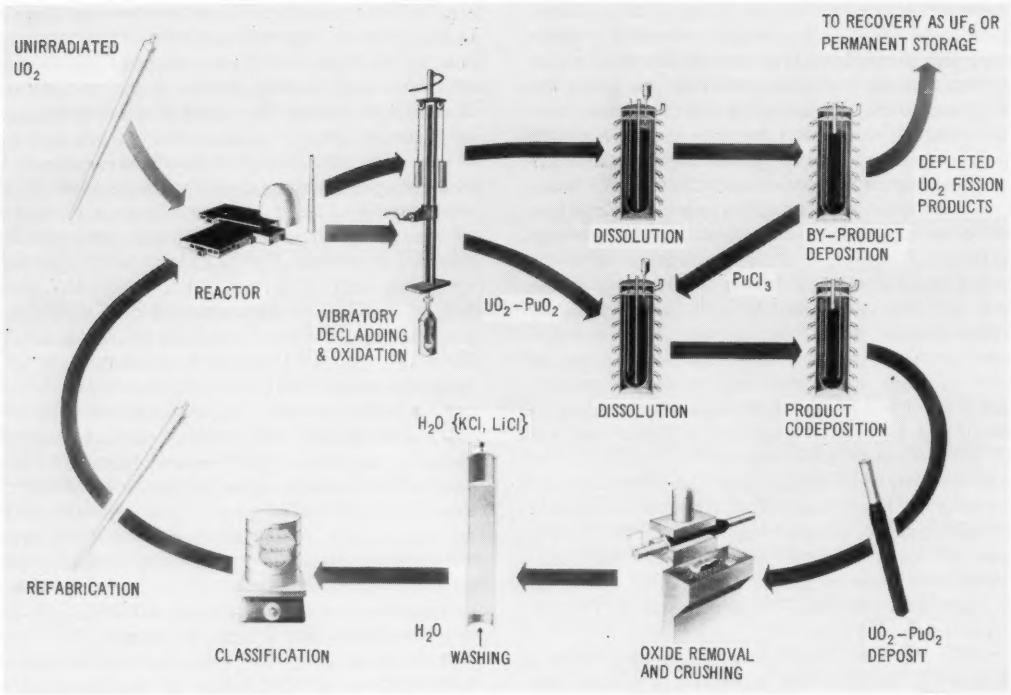


Fig. 3 Close-coupled plutonium recycle system.[13]

and iodine entrapment by a low-temperature chlorine liquefaction were described in an earlier report.[15,16]

The salt-cycle process was demonstrated[13] in remotely operated pilot-plant equipment in a high-level radiochemistry facility. The electrodeposition of UO_2–PuO_2 in 10- to 20-lb batches from molten equimolar LiCl–KCl was accomplished with feed material from 75 half-length sections of fuel rods that had been irradiated up to 5000 Mwd/metric ton in the Plutonium Recycle Test Reactor (PRTR).

The time required to declad cold-swaged half rods by mechanical vibration increased almost directly with the fuel burnup. Vibratory-compacted half rods, however, were declad in about 5 min for all cases up to an exposure level of 4300 Mwd/metric ton. Average waste losses were 0.6 and 0.25% for cold-swaged and vibratory-compacted half rods, respectively, at the higher burnups. These losses were greater than the 0.1% loss experienced[12] in cold pilot-plant tests.

The dissolution rates for the mixed-oxide feed material ranged from 0.05 to 0.5 lb of uranium per hour and were lower than expected from cold pilot-plant studies. The introduction of chlorine through a submerged annular gas lift, instead of the simple tubes used in initial tests, improved the dissolution rate in some cases but did not do so consistently. This was partially attributed to inconsistent particle-size reduction and incomplete conversion of the feed to U_3O_8 during the preceding oxidation step. The precipitation of a portion of the plutonium as PuO_2 at the gas–liquid interface on the walls of the quartz vessel during dissolution resulted in incomplete recovery of the plutonium. The volatilization of zirconium, ruthenium, and antimony from the salt during dissolution of the irradiated fuel was adequately handled by the in-cell off-gas treatment system.

In tests of the partition step, the degree to which the UO_2 was partitioned from the plutonium showed a strong dependence on the manner in which the salt bath was preconditioned prior to electrolysis. Plutonium decontamination factors of 120, 80, and 20 were obtained when the melt was preconditioned (1) by adding particulate graphite and sparging with chlorine gas, (2) by preelectrolysis, and (3) by sparging with hydrogen chloride gas, respectively. Each treatment was an attempt to shift the plutonium into the non-depositing Pu(III) state prior to electrolysis. Conditions for partition always became more favorable during the course of a partition electrolysis.

Controlling the enrichment of the PuO_2 in the UO_2 during codeposition was difficult. The problem was attributed to the formation of U(IV) that suppressed the formation of the codepositing plutonyl species.

The presence of U(IV) in the melt was an unpredictable function of the surface area of the particulate graphite introduced by corrosion of various pieces of graphite in the system. A method of measuring the PuO_2 enrichment and suitably controlling the electrolysis conditions was developed to control the plutonium enrichment of the deposited product.

Overall decontamination factors of approximately 3 for neutron-poisoning fission products were obtained during codeposition in the hot-cell, pilot-plant runs. The major difficulty was the removal of rare earths and the elements between rhodium and silver. These decontamination factors were lower than those obtained in the laboratory and were attributed to poorer process control and greater leakage of air during the hot runs.

The electrodeposited oxide was readily stripped from the pyrolytic graphite cathode without any apparent carbon contamination. Water washing of the oxide reduced the chloride content to less than 50 ppm in some cases. When the oxygen-to-uranium ratio of the material was too high, it was lowered to an acceptable level by hydrogen reduction.

A total of 13.75 kg of UO_2–2 wt.% PuO_2 codeposited in the pilot-plant equipment was vibratorily compacted into fuel rods, and 4.7 kg of the recycled material is undergoing irradiation in the PRTR.

Russian investigators have reported[17] a study of the decomposition of UO_2Cl_2 and its interaction with UO_2 in liquid NaCl–KCl.

METAL PREPARATION BY ELECTROLYSIS

Several recent reports are related to electrolytic procedures to prepare or purify uranium or plutonium metal. In one process[18] the electroreduction of uranium oxides was performed in a molten-fluoride-salt electrolyte at temperatures above 1130°C. In a single-phase liquid system, current efficiencies up to 44% were obtained, and the operation was quite flexible. When the surface of the salt melt was frozen, current efficiencies were about 80%, but the operation was limited in some respects.

A translation[19] of a Japanese report[20] describes an electrolytic refining procedure that produces uranium metal containing less than 100 ppm total impurities. Uranium tetrafluoride dissolved in liquid KCl–LiCl eutectic was reduced to metal and deposited on tungsten or uranium electrodes at voltages of 0.5 volt or less and at cathode-current densities of 0.33 amp/cm^2 or less. In general, at the lower voltages, the uranium crystals were larger and had lower metallic impurities but also had higher amounts of occluded salt. Although the decontamination from aluminum was small, other elements including carbon and nitrogen were removed to a level below the analytical limits. Occluded salt was removed by volatilization when the deposited uranium crystals were arc or electron-beam melted.

A U. S. patent[21] was issued for a process for the electrorefining of plutonium. Impure plutonium is melted with an electrolyte consisting of NaCl, KCl, and $MgCl_2$. After the two-phase liquid system is agitated, electrolysis is begun. Chemical reaction between $MgCl_2$ and plutonium metal provides sufficient plutonium ions in the salt phase to start the electrolysis.

Zone Melting

An investigation to establish the variables and parameters necessary to purify uranium by zone refining was reported.[22] With optimum cleaning procedure, molten-zone width, direction and rate of travel of the molten zone, length of zone-refined area, and vacuum, purified uranium was prepared with total impurities of less than 30 ppm. Analyses for 76 individual elements with estimated accuracies in the 10 to 100 ppb range were made with a spark-source mass spectrometer.

References

1. *Power Reactor Technol. Reactor Fuel Process.*, 10(1): 80 (Winter 1966–1967).
2. R. M. Adams and A. Glassner (Comps.), Reactor Development Program Progress Report, February 1967, USAEC Report ANL-7308, pp. 10-11, 35-39, Argonne National Laboratory, Mar. 28, 1967.
3. R. M. Adams and A. Glassner (Comps.), Reactor Development Program Progress Report, January 1967, USAEC Report ANL-7302, pp. 22, 54-55, Argonne National Laboratory, Feb. 24, 1967.
4. R. M. Adams and A. Glassner (Comps.), Reactor Development Program Progress Report, March 1967, USAEC Report ANL-7317, pp. 30-32, 59-61, Argonne National Laboratory, Apr. 28, 1967.
5. R. B. Briggs, Molten-Salt Reactor Program, Semiannual Progress Report for Period Ending August 31, 1966, USAEC Report ORNL-4037, pp. 142-158, Oak Ridge National Laboratory, January 1967.
6. F. F. Blankenship, Chemical Separations in Molten Fluorides, USAEC Report ORNL-P-2601, Oak Ridge National Laboratory, 1966.
7. N. R. Chellew and R. K. Steunenberg, Extraction of Plutonium from Uranium–Plutonium Alloy with Uranium Trichloride, *Nucl. Appl.* 3(3): 142-146 (March 1967).
8. R. C. Vogel, M. Levenson, J. H. Schraidt, and J. Royal, Chemical Engineering Division Research Highlights, May 1965–April 1966, USAEC Report ANL-7175, pp. 10-11, Argonne National Laboratory.
9. I. O. Winsch, M. L. Kyle, R. D. Pierce, and L. Burris, Jr., Tungsten Crucibles in Pyrochemical Processing of Nuclear Fuels, *Nucl. Appl.* 3(4): 245 (April 1967).
10. A. Schneider, J. van Impe, and W. Meyer-Jungnick, Method and Device for the Separation of Fission Products Formed After Insertion in a Nuclear Reactor, German Patent No. 1,220,616, to Leybold Hochvakuum Anlagen GmbH., July 7, 1966.
11. J. B. Knighton and R. K. Steunenberg, Purification of Molten Salts, U. S. Patent No. 3,271,133 to USAEC, Sept. 6, 1966.
12. G. Jansen, Jr., L. K. Mudge, and P. W. Smith, Engineering Development of the Salt-Cycle Process, USAEC Report BNWL-354, Battelle–Northwest, December 1966.
13. W. R. Bond, G. Jansen, Jr., and L. K. Mudge, Demonstration of the Salt-Cycle Process in a High-Level

Radiochemical Facility, USAEC Report BNWL-355, Battelle–Northwest, December 1966.
14. *Reactor Fuel Process.*, 8(1): 31 (Winter 1965); 8(2): 103 (Spring 1965); 8(3): 169 (Summer 1965); 9(2): 101 (Spring 1966).
15. L. K. Mudge and J. R. Lundquist, Chlorine Recovery and Radioiodine Behavior in the Salt-Cycle Process, USAEC Report BNWL-279, Battelle–Northwest, July 1966.
16. *Power Reactor Technol. Reactor Fuel Process.*, 10(2): 168 (Spring 1967).
17. M. V. Smirnov, V. E. Komarov, and A. P. Koryushin, Decomposition of UO_2Cl_2 and Its Interaction with UO_2 in Molten NaCl–KCl, *At. Energ. (USSR)*, 22(1): 30 (1967).
18. R. D. Piper, Laboratory Development of the Uranium Electroreduction Process, USAEC Report MCW-1500, Mallinckrodt Chemical Works, July 1, 1966.
19. K. Nakajima, M. Koizumi, M. Suzuki, and T. Kuroda, Production of High Purity Uranium by Fused Salt Electrolysis, JAERI Report NSJ-tr-72, November 1966.
20. K. Nakajima, M. Koizumi, M. Suzuki, and T. Kuroda, *Denki Kagaku*, 33(2): 122-126 (February 1965).
21. J. L. Long and R. D. Schweikandt, Electrorefining of Plutonium, U. S. Patent No. 3,282,806, to USAEC, Nov. 1, 1966.
22. J. L. Beiber, J. M. Schreyer, and E. L. Williams, Purification of Uranium by Zone-Refining Techniques, USAEC Report Y-1564, Union Carbide Corporation, Nuclear Division, Feb. 7, 1967.

Research and Development on Aqueous Processing

By C. E. Stevenson and D. M. Paige

Recent research and development activities related to the reprocessing of irradiated nuclear reactor fuels by aqueous solution methods include studies of means of dissolving Zr–U alloys, Zircaloy-clad ZrO_2–UO_2 ceramics, and the uranium content of graphite-matrix fuels. In the area of extraction processes, the recovery of actinides after nuclear detonations in volcanic formations is reviewed, an alternative neptunium extraction from Purex wastes is described, and a laboratory method used for ^{231}Pa purification is noted. Other topics include test and development of novel processes and equipment, and an evaluation of the performance and experience of processing plants using direct and remote maintenance techniques.

Preparation of Fuel for Processing

Zirconium–uranium alloy fuels may be dissolved without excessive corrosion in 304L stainless-steel vessels by a reagent composed of $2M$ HF–$1M$ HNO_3–$1M$ $Al(NO_3)_3$–$0.1M$ $(NH_4)_2Cr_2O_7$, with periodic additions to HF to attain a ratio of 5 moles of HF per atom of zirconium dissolved. Zircaloy-clad PuO_2–UO_2 fuels are declad in NH_4F–NH_4NO_3 solution, followed by dissolution in 5 to $11M$ HNO_3 containing a carefully limited amount of F^-. Zircaloy-clad ZrO_2–UO_2 fuel is declad with $5N$ HF, the ZrO_2 is leached from the compact with $20N$ HF, and the remaining uranium is dissolved in $0.1M$ chromic acid. Graphite-matrix fuels containing pyrolytic graphite-coated uranium dicarbide particles are prepared by grinding and leaching with HNO_3 or by disintegration and dissolution with H_2SO_4–HNO_3.

DISSOLUTION OF Zr–U ALLOY

A process for the dissolution of Zircaloy-clad Zr–U alloy (with minor concentrations of uranium) in 309SCb stainless-steel vessels has been developed at the Savannah River Laboratory (SRL).[1] The dissolving technique is based on the use of quite dilute ($1M$) HNO_3 at relatively low temperatures (35 to 60°C), with just enough HF (5 moles per atom of zirconium) to dissolve the zirconium. The HF is not initially present, but is added continuously during dissolution at a $3.0M$ concentration (with agitation), the addition rate being limited by heat removal and off-gas handling capacity. The presence of HNO_3 reduces the generation of hydrogen and avoids insoluble UF_4 by oxidizing the uranium. Control of acid concentrations limits the corrosion of the stainless steel to the order of mils per month of vessel use. In this way, plants with existing 309SCb stainless-steel dissolving equipment may be used for the processing of reactor fuels such as that of the Shippingport Pressurized Water Reactor (PWR) seed core. Clear and stable dissolver solutions, suitable by adjustment for tributyl phosphate (TBP) solvent extraction, can thus be prepared with $0.5M$ Zr, $0.4M$ NO_3^-, and $2.5M$ F^-.

In a later development of this process reported by Caracciolo and Rust,[2] consideration has been given to modifications appropriate for the use of dissolvers constructed of 304L stainless steel, as the 309SCb alloy became less available. The 304L stainless steel is too easily corroded for use for the process just described. For a mixture of $2M$ HF–$1M$ HNO_3, it was found that, by the addition of Al^{3+} [as $Al(NO_3)_3$] to a concentration of $1M$, corrosion could be markedly reduced while an adequate rate of dissolution of zirconium was maintained. By a further addition of $0.1M$ $(NH_4)_2Cr_2O_7$, corrosion was reduced eightfold with only a small reduction in the rate of dissolution of unoxidized Zircaloy, as shown in Table 1.

In this case the choice was to add HF intermittently rather than continuously to maintain an adequate dissolution rate without producing precipitation of either AlF_3 or ZrF_4. If ZrF_4 is produced, it is particularly difficult to redissolve. As shown in the stability plot given in Fig. 1, stepwise additions of HF at intervals of several hours can be used to bring the solution

composition through the region of stability, while the zirconium concentration is brought up to the desired value of 0.5*M*. Of course, the frequency of addition must be determined by the amount and nature of the low and continues to be low until pits and fissures in the film are formed, whereas unoxidized Zircaloy commences to dissolve immediately at a relatively high rate. With oxidized Zircaloy an induction period of the order of 4 to 8 hr is observed, and the addition of HF is not required until this point is well past.

Table 1 EFFECT OF ADDITIVES ON ZIRCONIUM DISSOLUTION AND STAINLESS-STEEL CORROSION RATES[2]

Dissolving reagent	Zircaloy-2 dissolution rate,* mils/hr		304L S.S. corrosion rate in liquid, mils/year		Ratio of Zr/S.S. penetration rates	
	50°C	90°C	50°C	90°C	50°C	90°C
2*M* HF, 1*M* HNO_3	128	367	>5,000	>5,000	<300	<300
2*M* HF, 1*M* HNO_3, 1*M* $Al(NO_3)_3$	10	27	16	212	5,500	1,100
2*M* HF, 1*M* HNO_3, 1*M* $Al(NO_3)_3$, 0.1*M* $(NH_4)_2Cr_2O_7$	7	20	2	22	31,000	8,000

*Nonoxidized.

Zircaloy surface and is markedly affected by the presence of adherent oxide deposits.

Normally, Zircaloy fuel elements are tested for defects before irradiation by steam autoclaving, and in this process a tenacious film of ZrO_2 is formed. The initial rate of attack by $HF-HNO_3$ mixtures is

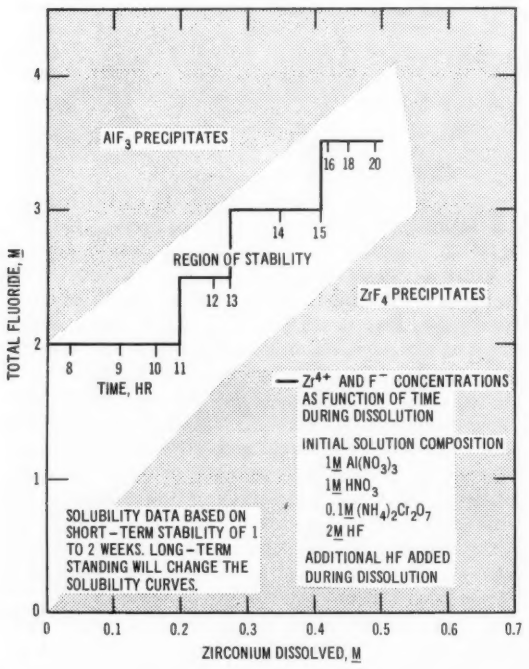


Fig. 1 Stability of dissolver solution[2] at 70°C. (Reprinted from Industrial and Engineering Chemistry, Process Design and Development, 5(4): 365 (October 1966). Copyright 1966 by the American Chemical Society. Reprinted by permission of the copyright owner.)

Normally, Zircaloy fuel elements are tested for defects before irradiation by steam autoclaving, and in this process a tenacious film of ZrO_2 is formed. The initial rate of attack by $HF-HNO_3$ mixtures is

Because of the presence of Al^{3+}, the dissolver solution may be used without further adjustment for extraction of uranium. Such processing must be accomplished shortly after dissolution, however, since zirconium and aluminum may gradually precipitate over a period of several weeks.

At SRL a satisfactory test of this process was accomplished by the dissolution of more than 99.5% of a section of a plate-type PWR seed element (6.3% U–93.7% Zircaloy-2), weighing 36 lb, in 24 hr at 68 to 75°C. Corrosion of the 304L stainless steel was at rates of 10 to 15 mils/year in the liquid phase and <1 mil/year in the vapor phase.

DISSOLUTION OF PLUTONIUM RECYCLE TEST REACTOR FUELS

A procedure for the dissolution of Zircaloy-clad Pu–Al alloy fuel used in the Plutonium Recycle Test Reactor (PRTR) was recently described in *Reactor Fuel Processing*,[3] and the development of methods for handling similarly clad PuO_2-UO_2 fuels for this reactor had been discussed previously.[4] Schulz of Battelle–Northwest has reported recent experience with the latter fuels.[5] The typical fuel element is a 0.57-in.-OD Zircaloy tube (35-mil wall thickness) that contains vibrationally compacted or swage-compacted UO_2 with 0.5 or 1.0% PuO_2 and is prepared either by mixing powders or by coprecipitation. The cladding is readily removed by boiling with 4.0*M* $NH_4F-0.36M$ NH_4NO_3, as used in the Hanford Redox-plant dissolver, or in 5.7*M* $NH_4F-0.55M$ NH_4NO_3, as proposed for another dissolver. However, considerable conversion of the UO_2 to UF_4 occurred, which is of importance since the fluoride produced will accelerate corrosion of stainless steel during subsequent dissolution of the core in HNO_3. It is, of course, important to remove the decladding solution as

completely as possible. Uranium and plutonium losses to the decladding solution were of the order of 1% or less.

For the subsequent core dissolution, three procedures were compared: (1) dissolution for 2 to 4 hr at 80°C in 11*M* HNO_3–0.1*M* NH_4F; (2) dissolution for 2 to 4 hr in boiling 5*M* HNO_3–0.25*M* NH_4F–0.75*M* $Al(NO_3)_3$, followed by treatment of any residue with 10*M* HNO_3–0.1*M* NH_4F at 80°C; and (3) dissolution for 3 to 7 hr in boiling 8.7*M* HNO_3–0.15*M* NH_4F. Tests were conducted with these reagents both with PuO_2 powder and with the PuO_2–UO_2 fuel, mechanically or chemically declad. The first two procedures appeared to be adequate for use in stainless-steel dissolvers without excessive corrosion, and the single-step process is recommended as the more suitable assuming that formation of UF_4 during decladding is minimal. The second procedure may offer more protection if large amounts of UF_4 are formed. The third procedure is only suitable for use in a fluoride-resistant dissolver, such as one constructed of Nionel or an experimental alloy described as HAPO 20 (50% Ni, 25% Cr, 6% Mo, 16% Fe, 1% Ti, 1% Cu, 0.6% Mn, 0.4% Si). A recommended limit for the fluoride content of the dissolver solution in such a case is 0.25*M*.

The PuO_2 content of unirradiated fuel produced by mixing PuO_2 and UO_2 powders is not readily dissolved by HNO_3, and the presence of fluoride appears to be necessary for rapid dissolution of PuO_2. However, PuO_2 in the form of coprecipitated, ignited, and hydrogen-reduced oxides appears to be in solid solution and dissolves along with the UO_2. Similarly, irradiation of the mixed oxides facilitates dissolution of PuO_2, and PuO_2 in a sample irradiated to 5000 Mwd per ton of uranium dissolved completely in 2 hr in boiling 11*M* HNO_3 under conditions where less than 1% of the PuO_2 in an unirradiated sample was dissolved.

In three Redox-plant tests of dissolving irradiated UO_2–PuO_2 fuels (290 to 11,400 Mwd/ton), the decladding step resulted in 0.44 to 0.72% loss of uranium and 0.22 to 0.46% loss of plutonium. Core dissolution in 11.9*M* HNO_3–0.1*M* NH_4F was carried on for 7 to 9 hr and yielded solutions containing 7.4 to 8.7*M* HNO_3, 0.5 to 0.6*M* $UO_2(NO_3)_2$, 0.3 to 0.8*M* F^-, and 0.03 to 0.04*M* Zr. The higher-than-desirable fluoride concentrations were attributed to carry-over of heels of decladding waste solution.

DISSOLUTION OF ZIRCALOY-CLAD ZrO_2–UO_2 FUEL

A recent seed fuel for the Shippingport PWR, which utilized fuels in seed-blanket configurations, is composed of ZrO_2–UO_2 compacts (28 wt.% UO_2) in wafer form, clad in Zircaloy-4, such that the Zircaloy comprises about 78% of the fuel element. This is identified as Seed 1 of Core 2. Preliminary development of a process for dissolving this fuel in Monel equipment available at the Idaho Chemical Processing Plant (ICPP) was discussed previously in *Reactor Fuel Processing*.[6] The proposed process consisted in dissolving the cladding in dilute HF below the boiling point, removing the cladding solution, dissolving the ZrO_2 in boiling concentrated HF while the dissolution of uranium is inhibited by the addition of KF to form the insoluble double salt with UF_4, removing the zirconium-containing solution, and dissolving the residue in dilute chromic acid. By the addition of HNO_3 and a small amount of $Al(NO_3)_3$, uranium may be extracted from the latter solution with TBP. The basis of the process lies in the need to use HF for dissolving both zirconium and ZrO_2 and in the very low rate of dissolution of the latter compared to the former (by a factor of 10^3).

Butzman and Newby have recently published a report[7] on this process, in which the flow sheet is essentially as described earlier. The flow sheet shown in Fig. 2 indicates the steps and quantities needed for the decladding and dissolution of 1 kg of fuel.

In the decladding step with 5*N* HF–0.1*M* KF, a very slow attack on the ZrO_2–UO_2 compacts occurs. However, only 15 min is required to dissolve the Zircaloy, and by limiting the decladding period and holding the temperature to the order of 70°C, the losses of uranium at this stage are low and are further reduced by the addition of KF to the order of a few hundredths of 1%. The ZrO_2 dissolution step with concentrated acid requires several hours, and the acid strength must stay at a high level to sustain a reasonable rate of dissolution. Thus only a small fraction of the acid is utilized, and the zirconium content achieved is only 0.39*M*. The thin layer of pyrolytic carbon that coats the compacts is disintegrated in this process, and the wafers are converted to porous $UF_4 \cdot {}^3/_4H_2O$ with their form substantially preserved. Again, the presence of 0.05*M* KF should limit the uranium loss to the zirconium leach to a few hundredths of 1%. However, in a single test in dissolving irradiated fuel, the combined decladding–leaching uranium loss was 0.62%. The cause of the higher loss in this case has not been explained. Boric acid may be added to either or both reagents for criticality control without interfering with dissolution. The decladding and the wafer-dissolution wastes may be combined for calcination with lime, or they may advantageously be separately treated since the decladding waste will presumably be less radioactive.

The dissolution of the $UF_4 \cdot {}^3/_4H_2O$ residue is readily accomplished by boiling in 0.1*M* chromic acid, with or without 0.37*M* boric acid. Chromic acid produces U(VI) suitable for extraction. It is proposed to dilute the resulting solution with an equal volume of 6*N* HNO_3, containing 0.4*M* $Al(NO_3)_3$ to complex F^- accompanying the uranium, in order to provide

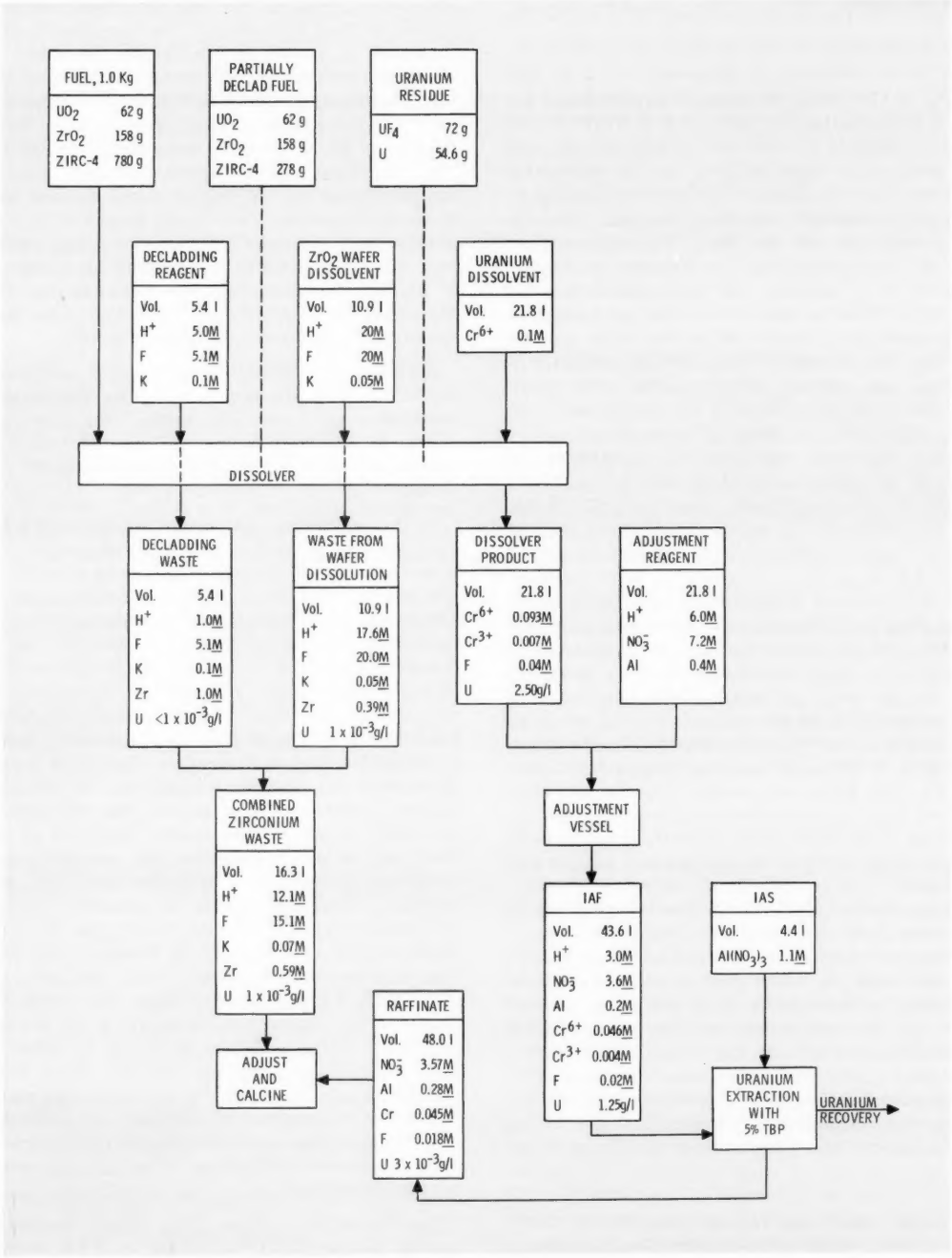


Fig. 2 Reference flow sheet for PWR Core 2 Seed 1 fuel.[7]

suitable salting strength for extraction of the uranium with 5% TBP in a hydrocarbon diluent.

Corrosion of Monel equipment in this processing occurs at similar rates to those observed for the HF process for U–Zr alloy. In a laboratory-scale dissolver, corrosion rates of 4 to 18 mils/month were observed for the ZrO_2–UO_2 process compared to a long-term corrosion rate of 5.8 mils/month observed in the plant dissolver operating on U–Zr fuels.

A PWR seed fuel has been proposed which would consist of similar compacts of ZrO_2–UO_2 containing 2% CaO. In an attempt to dissolve ZrO_2 from declad wafers of this composition, the rate of dissolution of ZrO_2 was markedly reduced (by a factor of 10) as compared to the behavior of wafers not containing CaO, and dissolution was incomplete. Consequently this process may not be practical for ZrO_2–UO_2 containing 2% CaO.

PREPARATION OF GRAPHITE FUELS

The processing of nuclear fuels composed primarily of graphite-containing fissionable material in the form of decomposed salts or of coated or uncoated carbide particles is rendered difficult because of the relatively insoluble and refractory nature of the graphite matrix. A number of methods have been proposed for preparing such fuels for uranium recovery. Currently, the most attractive method is combustion of the fuel in a fluidized bed. This method and an alternative method of reacting with HNO_3 in the liquid phase at 300°C and 1500 to 2000 psi were discussed previously in *Reactor Fuel Processing*.[8] Work on a two-stage fluidized-bed combustion technique is proceeding at the ICPP.[9] Other methods have included grinding and HNO_3 leaching, electrolytic dissolution in HNO_3, and mixed-acid ($H_2SO_4 \cdot HNO_3$) dissolution, which have also been reviewed in *Reactor Fuel Processing*.[10,11] Decker and Hammer have recently reported work at ICPP[12] indicating that, although the grind-leach and mixed-acid oxidation processes are feasible, they offer no real advantages over the fluid-bed combustion process and that the electrolytic dissolution technique does not give adequate uranium recovery.

Tests were conducted on fuel elements composed of pyrolytic graphite-coated uranium dicarbide particles dispersed in a graphite matrix, with NbC coolant channels. These elements were made for the Kiwi nuclear rocket reactor but are similar to those used in the Peach Bottom and proposed Fort St. Vrain HTGR's. When fuel pieces in contact with a platinum anode in 70% HNO_3 were subjected to potentials of 8 to 33 volts at 47 to 98°C, the graphite disintegrated rapidly, exposing the coated fuel particles. However, the coating was penetrated only slowly, or not at all, and dissolution of uranium in 8 hr ranged from 1 to 24%. With the addition of 0.4*M* $K_2Cr_2O_7$, 39% of the uranium was dissolved, but in no case was adequate recovery obtained.

Adequate recovery of uranium by HNO_3 leaching from fuel prepared by grinding requires that size reduction be effected by producing particles smaller than the smallest coated uranium carbide particles. From such material, 15.7*M* HNO_3 leached >99% of the uranium present in 3 hr. For continued processing by solvent extraction, clarification of the feed appears to be desirable to avoid emulsification problems, although extraction of uranium from a graphite slurry (30 g of graphite per liter) was feasible with the addition of 200 ppm gelatin to the slurry.

Treatment of graphite fuel with hot H_2SO_4–HNO_3–H_2O mixtures rapidly disintegrates the graphite matrix, but several hours' boiling with the mixture is required to penetrate the pyrolytic carbon coating. Solutions containing 22 to 85 mole % H_2SO_4, 7 to 27 mole % HNO_3, and 0 to 60 mole % H_2O effected rapid disintegration, and high uranium recovery (in 16 hr) was obtained with the following mixtures:

(1) 85 mole % H_2SO_4–15 mole % HNO_3; 99% U recovery
(2) 44.5 mole % H_2SO_4–12.5 mole % HNO_3–43 mole % H_2O; 99.9% U recovery

It would be expected that high recoveries would be obtained anywhere within the range of 44 to 85 mole % H_2SO_4. The mixture can be contained satisfactorily in tantalum (corrosion loss < 0.02 mil/month at 120°C) and possibly in Duriron, although, in a 30-day test in which HNO_3 may have been lost, the latter material was badly corroded. A proposed flow sheet for this process, including preparation of a suitable solvent extraction feed by addition of $Al(NO_3)_3$ solution, is given in Fig. 3. A dilution factor of 27 is necessary to lower the sulfate concentration to 0.5*M* and permit adequate extraction.

Solvent Extraction Processes

The concentration and purification of actinide elements produced by a nuclear detonation in volcanic formations may be effected by solvent extraction following removal of SiO_2 from the finely ground material by volatilization of SiF_4 with HF. Trilaurylamine is an efficient solvent for extraction of neptunium from Purex-process wastes. Protactinium may be purified by diisobutyl ketone and diisobutylcarbinol extraction.

ACTINIDE RECOVERY FROM NUCLEAR DETONATIONS IN VOLCANIC TUFF

In connection with the Plowshare Program, consideration has been given to the production of transplutonium isotopes by underground detonation of thermonuclear devices. The extremely high neutron flux (>10^{26} neutrons/cm^2) that is available leads to successive neutron capture and the formation of high-atomic-mass elements. Recovery of the isotopes

in quantity will then require large-scale novel chemical-processing operations. Such operations can be simplified by carrying out the detonation in a salt formation since common salt is readily dissolved in water. Karraker and Perkins[13] have described a proposed process for separation of salt, dissolving the residue, and extracting and concentrating the transplutonium elements with TBP.

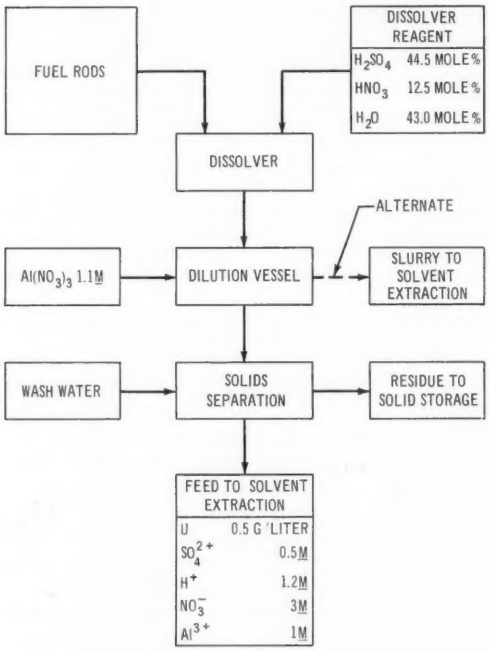


Fig. 3 Mixed acid flow sheet for graphite fuel.[12]

Since a new test site might be required to exploit salt formations in this manner, the same authors have considered more recently the more difficult problem of recovering transplutonium elements from volcanic formations such as occur at the Nevada Test Site (NTS).[14] The characteristic formations in which detonations are produced at that location consist of tuff, a porous volcanic rock, and alluvium, a sedimentary deposit produced by the weathering of tuff. Both are alkali-metal aluminum silicates, consisting of 61 to 65% SiO_2, 11 to 13% Al_2O_3, 4.5 to 6.9% $K_2O + Na_2O$, other metals in minor quantities, and 13 to 20% H_2O. Transplutonium isotopes are presumably embedded in the partly fused masses.

The large quantity of SiO_2 is obviously the source of the principal problem in chemical processing since dissolution of the bulk is necessary to effect the desired recovery. Attack on the separation of SiO_2 by HF volatilization, by Na_2CO_3 fusion and HNO_3 precipitation, or by strong acid leaching were considered; treatment with gaseous HF was chosen for further development owing to simplicity of equipment required and relative ease of scale-up.

From laboratory-scale studies it was found that crushing the mined debris from a detonation to −30 mesh and volatilization of SiF_4 by treating the crushed material with a 100% excess of anhydrous HF gas at 500 to 550°C for 15 to 20 min converted the silicates to metal fluorides, largely soluble (93 to 99%) in $1M$ $Al(NO_3)_3 - 1M$ HNO_3. The bulk of the actinides should be contained in this solution. Grinding the mined debris to finer particles (−50 or −100 mesh) slightly decreased the amount of residue and the insoluble fraction, particularly when working with alluvium. About 95% of the SiO_2 could be removed, and a vertical flow of gas through a fixed bed was more effective than a horizontal flow of gas over the bed surface. A temperature of at least 500°C was required, but higher temperatures did not reduce the insoluble residue significantly.

The choice of dissolving reagents for the residue left after SiO_2 removal was made on the basis of providing a solution suitable for solvent extraction by TBP, and a nitrate system was selected; $6N$ HNO_3 was much less effective than less concentrated acid containing $Al(NO_3)_3$. By complexing F^- ions the latter also reduces equipment corrosion. The undissolved solids could not be completely separated by centrifugation since SiO_2 was present in colloidal form. A minimum ratio of 10 ml of dissolving reagent per gram of the residue from HF treatment was found necessary for good solids separation. Decantation followed by gelatin treatment was effective in preparing a feed suitable for solvent extraction by TBP. The process recommended is shown in Fig. 4.

The proposed solvent extraction process consists of two cycles of multistage extraction with 30% TBP in a hydrocarbon diluent, with evaporation of the product from each cycle. The flow sheet is very similar to that given for recovery from salt in Ref. 13. The feed-to-strip ratio in each cycle is about 16/1; the overall concentration factor is better than 5000 from feed to final evaporated product.

The overall process was demonstrated using 30 g of tuff recovered from a detonation at the NTS. Hydrofluorination of the −30 + 50 mesh material was carried out for 2 hr at 500 to 600°C to yield 6.5% solids not soluble in $1M$ $HNO_3 - 1M$ $Al(NO_3)_3$. The resulting slurry was decanted after settling, and the resulting solution was evaporated, partially neutralized, treated with gelatin (100 ppm for 2 hr at 70°C), and filtered. The feed for extraction with TBP contained 89% of the gross alpha activity in the tuff debris and 87% of the curium content. Batch extraction tests indicated that >99% of the ^{244}Cm should be recovered.

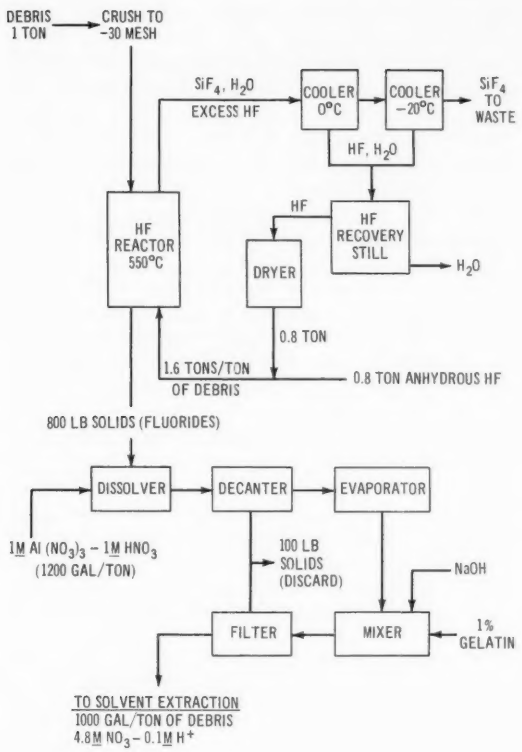


Fig. 4 Treatment flowsheet for recovery of actinides from tuff and alluvium debris.[14]

TRILAURYLAMINE EXTRACTION OF NEPTUNIUM

The recovery of neptunium as a by-product of the processing of plutonium-production reactor fuels is extensively practiced since the ^{237}Np secured is an excellent source for the production of ^{238}Pu useful for remote power supplies in space and other applications. A summary of Hanford and Savannah River Plant processes which appeared in *Reactor Fuel Processing*[15] indicated that the technique applied at the Hanford Purex plant consisted in carrying about 85% of the neptunium along with uranium into the second uranium decontamination cycle (after plutonium separation), and rejecting neptunium into the second-cycle aqueous waste (2DW) when uranium is extracted by the solvent. About two-thirds of the neptunium-bearing waste stream is recycled to the primary extraction (HA) column, and the remainder is diverted to a neptunium-accumulation extraction cycle (3A and 3B columns) using a TBP solvent, from which it is removed periodically for ion-exchange purification. Schulz of Battelle–Northwest has proposed[16] an alternative to this scheme in which the neptunium is forced into the primary extraction (HA) column aqueous waste stream by maintaining a high saturation of solvent with uranium, and adding $NaNO_2$ to establish and maintain the inextractable Np(V) state. Neptunium would be recovered from the HA waste by reduction to Np(IV) and extraction with diluted trilaurylamine (TLA), for further processing in the TBP accumulation and ion-exchange cycles. A comparison of the steps in the two processes is given in Fig. 5. The claimed advantages of the TLA scheme include increased flexibility and improved decontamination in the primary Purex operation, flexibility in waste rework to recover product losses, and greater overall recovery of neptunium and plutonium.

The applicability of TLA for neptunium and plutonium recovery has been described previously in *Reactor Fuel Processing*[17] in discussing French work on this subject. Its basis is the great selectivity of TLA for Np(IV) from nitrate media. The form in

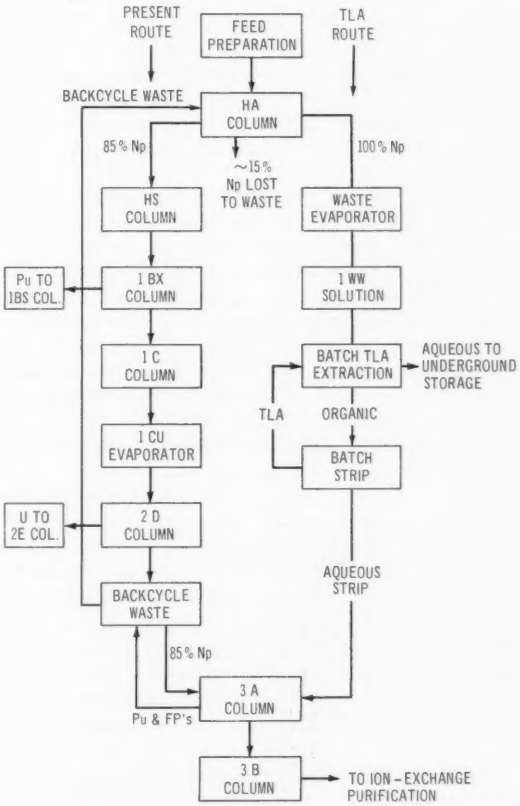


Fig. 5 Hanford Purex plant neptunium recovery schemes.[16] *Reprinted from* Industrial and Engineering Chemistry, Process Design and Development, *6(1): 115 (January 1967). Copyright 1966 by the American Chemical Society. Reprinted by permission of the copyright owner.*

which neptunium is extracted is presumably as an adduct formed as follows:

$$2(R_3NH^+NO_3^-) + Np(NO_3)_4 = (R_3NH)_2Np(NO_3)_6$$

In the present application, neptunium is contained in the waste solution (6.3*M* HNO_3, 0.35*M* Fe^{3+}, 0.7*M* Na^+, 0.8*M* SO_4^{2-}) in the Np(IV) and Np(V) states, and plutonium is in the Pu(IV) state. After reduction to Np(IV) by the addition of N_2H_4, to a concentration of 0.02 to 0.05*M*, 97 to 99% of the neptunium and plutonium are batch extracted by contacting 30 min with an equal volume of 0.3*M* TLA in a paraffinic diluent such as Soltrol 170. The neptunium and plutonium are stripped from the separated organic phase by contacting 30 min with an equal volume of 0.05*M* $(NH_2OH)_2 \cdot H_2SO_4$ at 50°C to provide a feed for TBP process recovery and purification as at present. Such an additional decontamination step is necessary because Ru, Ce, and Zr accompany Np through the TLA process in significant amounts. Carry-over of TLA into the TBP process adversely affects the decontamination from ruthenium effected in the Purex process, but as much as 5 vol.% TLA in the TBP extractant does not adversely affect uranium or plutonium extraction or decontamination from $^{95}Zr-^{95}Nb$. It was felt that the effect on ruthenium decontamination could be offset readily in other steps in the process.

Both chemical and radiolytic degradation of the TLA solvent may significantly affect the process. In contact with the feed solution, it was estimated that the exposure rate of the 0.3*M* TLA extractant is about 10^6 rads/hr. The effect of irradiation on neptunium distribution ratio is less in the presence of an aqueous phase than in its absence, but the Np(IV) distribution ratio decreased from 25 to 9 as the dose was increased to 84×10^6 rads. Small amounts of solids were precipitated, both by prolonged irradiation in paraffinic diluents and by long contact with nonradioactive acid waste.

DEGRADATION PRODUCTS OF TERTIARY AMINES IN THE CURIUM-RECOVERY PROCESS

In related work at Savannah River, the degradation of a tertiary amine solvent (*n*-octyl and *n*-decyl tertiary amines) was studied by West and Narvaez[18] for conditions applicable to a process for curium recovery. This process, as described in *Reactor Fuel Processing*,[19] first utilizes the tertiary amine solvent to convert from a nitrate system to a chloride system and utilizes the same type of solvent in two additional cycles to concentrate and purify the transplutonium elements. Process performance at high concentrations of ^{242}Cm (20 to 440 curies/liter) was noted earlier.[20] Slow loss of acid was noted in the nitrate-to-chloride conversion. Difficulties attributed to radiation effects resulted in poor cerium decontamination and in loss of acid in the decontamination cycles.

Narvaez and West attempted to isolate and identify degradation products of the solvent and diluent produced under conditions simulating the two extraction steps. For the conversion step, 30 vol.% tertiary amine in diethylbenzene (DEB) was mixed for 48 hr with an equal volume of 1.5*M* $Al(NO_3)_3-4.5M$ $LiNO_3-0.2M$ HNO_3 solution containing curium and cerium sufficient to provide a radiation dose of 26 watt-hr/liter, whereas for the decontamination step a similar solvent was agitated 3 hr with 10.5*M* $LiCl-0.07M$ $HCl-0.05M$ $SnCl_2$ containing sufficient curium to provide a dose of 7 watt-hr/liter. Gas chromatography, thin-layer chromatography, and infrared spectrometry were used for separation and characterization. Primary and secondary amines from the tertiary amine extractant, and aromatic alcohols and ketones from DEB, were the principal substances found. The tertiary amines were more susceptible to degradation in the chloride media, and DEB was more susceptible in the conversion step. Addition of 0.2*M* hydrazine in the latter step reduced the degradation of DEB from 0.8 to ~0.08% per watt-hour per liter, and no tertiary amine degradation was detected.

RECOVERY AND PURIFICATION OF ^{231}Pa

The chemistry of protactinium is of interest in connection with the development of the thorium fuel cycle in respect to providing required materials and techniques to obtain data needed for its adequate evaluation. Procedures for the recovery of ^{231}Pa from irradiated thorium have been described.[21] Brown, Dixon, and Jones at Harwell have recently described[22] a solvent extraction method for the purification of ^{231}Pa which, though it is primarily intended for the recovery of material used in laboratory studies, may have application to processes for protactinium recovery.

The method is based on dissolution in 9*M* $H_2SO_4-0.5M$ HF, addition of HCl and H_3BO_3, and extraction with diisobutylketone. Extraction with diisobutylcarbinol in kerosene from an HCl solution accomplishes further purification. The loss of Pa was <2%, and Sn, Pb, Cr, Zr, Ti, and Ta were removed to limits that were not spectrographically detectable, and good separation of Si, Al, Fe, and Mg from Pa was also accomplished.

Plant Design and Equipment Development

SEPARATIONS-PLANT MAINTENANCE

Maintenance of nuclear fuel chemical reprocessing plants has evolved over the years into two general methods: (1) contact maintenance and (2) remote maintenance. A good example of the latter method is to be found in the maintenance of the Hanford Purex plant, which is reported[23] by McIntosh and Raab.

This report covers the past 10 years of operation of the Purex plant. The basic maintenance rule followed by this plant is the immediate replacement of failed equipment; this has provided a 90% onstream operation.

Figure 6 shows relative cost accumulations on major equipment types for the last 10 years. Rotary

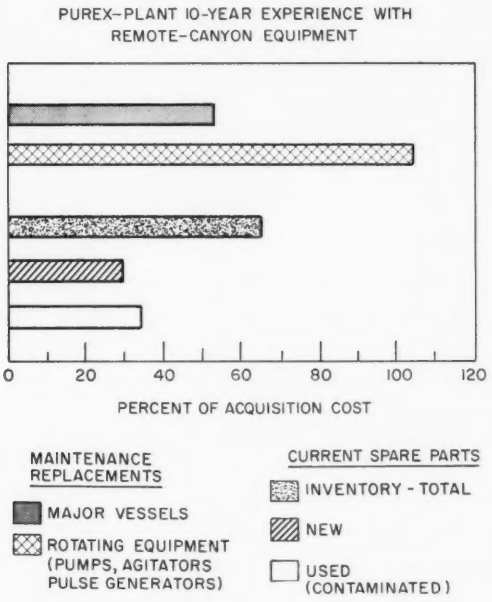


Fig. 6 Canyon equipment compared to original cost.[23]

equipment has experienced the highest replacement rate, but the authors report that a good percentage of these items were decontaminated and repaired for reuse. Table 2 shows costs and number of replacements for various individual items.

Table 2 CANYON EQUIPMENT REPLACED[23]

	Individual piece acquisition cost (average), $	Present complement	10-year failures
Pumps	7,000	22	60
Agitators	8,500	29	18
Pulse generators	35,000	11 (orig. 10)	5
Extraction columns	75,000	11 (orig. 10)	4
Concentrators	250,000	4 (orig. 5)	6
Off-gas heaters	26,000	4	12
Condensers	52,000	5 (orig. 6)	10
Tube bundles	50,000	8 (orig. 10)	42
Filters	26,000	4	4
Silver reactor	52,000	4	2
Dissolver tower	162,000	3	1
Acid rectifier	188,000	1	1
Air-operated valves	1,500	103	42
Rotameters	1,750	24	11
Jumper heads	300	2384	180

The basic tool used to accomplish these replacements is the 40-ton crane (two per canyon). The operator works in a shielded cab using a periscope optical system for viewing. Figure 7 shows an overall view of the crane working over the equipment canyon. The use of such a system limits the placement of vessels or groups of vessels to only one level since piping (jumpers), in general, must be attached from the top using special remote connector flanges. In addition, crane-operator skill is extremely important, and the authors mention that some of the men have had up to 20 years of experience in the operation of these cranes. New men require at least 2 years to gain the necessary experience, even if they are skilled at operating conventional cranes.

To work well, this maintenance system requires a very large inventory of spare equipment (Table 2). New equipment must undergo very rigid inspection and be "mocked up" before leaving the shop to be sure of a nearly perfect fit when installed. Nozzle dimensions are held to $\pm 1/16$ in. even on very large vessels.

Finally, the replaced equipment must be disposed of, and this is no small item for a plant as large as the Purex plant. The most desirable approach is to decontaminate and repair the equipment for reuse, but this is not always possible, and the equipment must be buried. Some items are placed in concrete boxes for burial in a trench at a cost of $5000 to $10,000 per box. A more interesting method for large equipment is railroad-tunnel burial. Figure 8 shows a partially completed tunnel. Large contaminated equipment is placed on a flatcar and pushed into the tunnel for indefinite storage. Costs are given as >$800/ft or ≈$36,000 per flatcar.

As a contrast to remote maintenance, experience using a contact-maintenance concept over the past 15 years at ICPP was recently discussed by Buckham.[24] As he points out, the main advantage of contact maintenance is that equipment can be stacked and hence much smaller high-cost shielded cells are required. The one disadvantage, of course, is that failure of an item in the process may shut down the entire line for an extended period. An example is mentioned where mechanical failure extended the process schedule for a campaign by 30%. Several ideas are mentioned to improve this, such as installed spare equipment, more reliable equipment, redesign for simpler equipment (e.g., air pulsers for columns vs. mechanical diaphragm pumps), and faster methods of decontamination when repair is required.

In general, experience at the ICPP has shown the direct maintenance principle to be practical, provided proper design is achieved. It can be expected that 80% on-stream factors are within reason for a plant, utilizing contact maintenance.

In general, as has been stated before, the remote-maintenance principle seems to be adapted to large

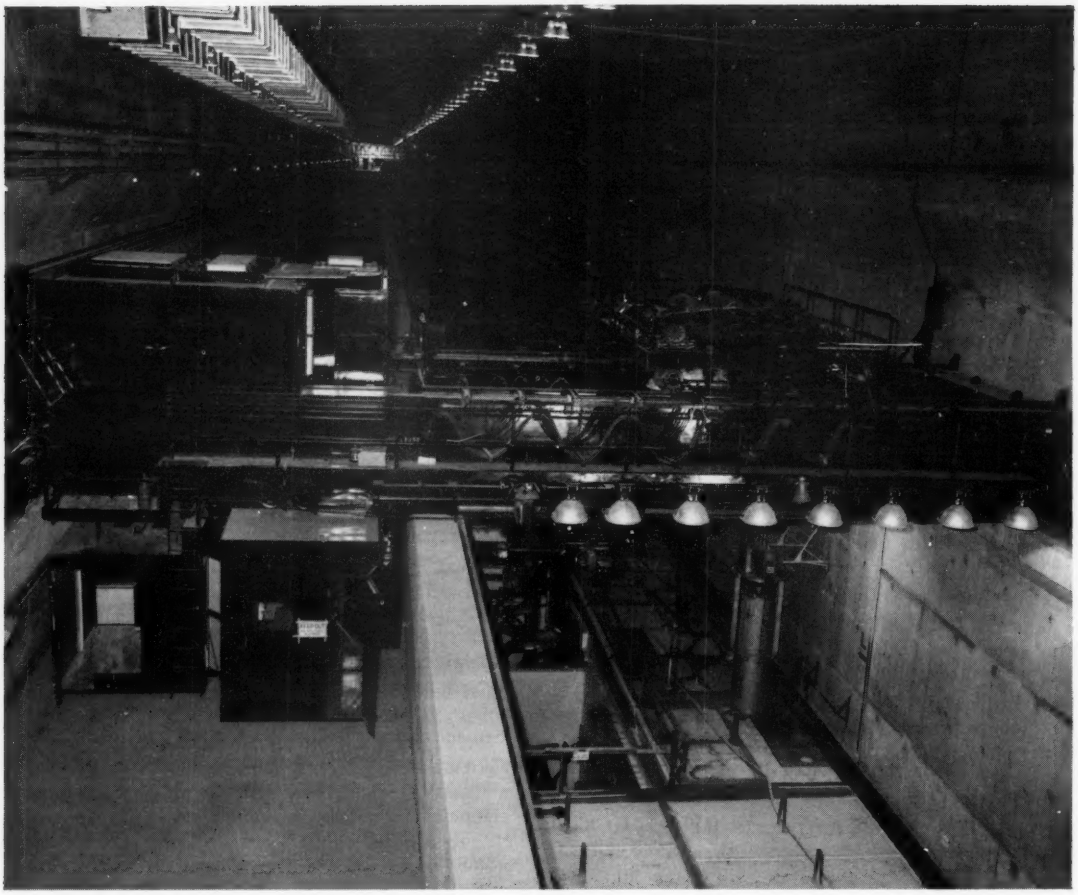


Fig. 7 Purex shielded crane.[23]

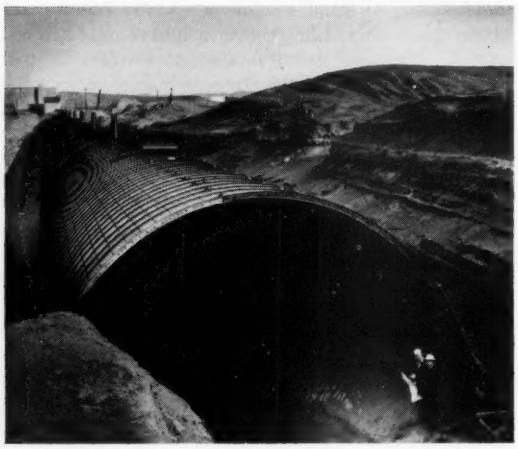


Fig. 8 Railroad spur burial tunnel.[23]

process plants and the direct-maintenance principle to smaller plants.

CONTINUOUS DISSOLVER

Recently, several special dissolver designs have been reviewed.[25] Lorenzen reported[26] on another special dissolver developed at Hanford for dissolution of plutonium-containing scrap. An average composition of the scrap is shown in the table below:

Material	Approximate wt.%
MgO crucibles (crushed)	20
MgO sand	27
CaF_2	40
CaI_2	2
Ca (metal)	8
$MgSiO_3$	<1
Pu (metal, oxides, and fluorides)	2 to 3

Nitric acid containing aluminum nitrate is used as the dissolvent. Some HF is produced from the CaF_2 reaction and is desirable for some of the reaction sequences, but does pose a corrosion problem to the dissolver.

Although this scrap-recovery equipment has been in operation since 1955, the batch dissolvers (440-liter tanks) have been considered undesirable mainly because of inadequate capacity. In addition, there was the possibility of a criticality incident as well as an explosion hazard from the hydrogen generated.

A dissolver of new design was conceived, based on the following criteria:

1. It must have sufficient capacity for processing the scrap material from current production, as well as being capable of working off the backlogged material.
2. It must be safe from a nuclear excursion standpoint, that is, geometrically favorable.
3. Sufficient freeboard is required for containment of the violent reaction between calcium metal and nitric acid.
4. Safe disposition of the highly explosive hydrogen gas generated by the dissolution of calcium metal is essential.
5. It must successfully resist the corrosive hot nitric-hydrofluoric acid media.

After several designs and prototype dissolvers had been evaluated, a successful continuous dissolver (Fig. 9) was put into operation. This dissolver incorporates the following features:

1. Nuclear excursions are avoided by geometrically favorable shape (a 6-in.-ID vapor section and a 4-in.-ID liquid section).
2. Two valves are used for charging solids.
3. A recirculating leg is used for efficient use of dissolvent.
4. A steam jacket supplies heat.
5. Inert gas sparge is used to dilute the hydrogen below the explosive limit.
6. Steam sparge is used to keep solids suspended.
7. A means of removing the undissolved solids without dismantling the dissolver is provided.

The report includes details on several of the intermediate designs, as well as on the final dissolver design.

The author reports[26] that the use of the newly designed dissolver (Fig. 9) and two slightly redesigned prototypes produced a threefold overall increase in dissolver capacity.

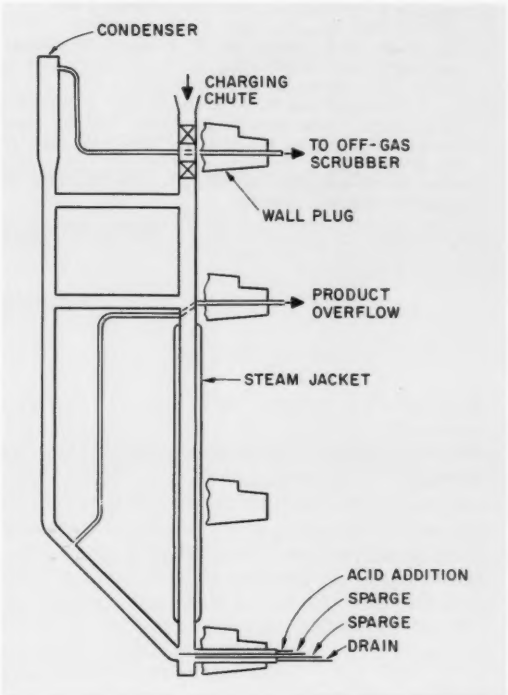


Fig. 9 Final design of third dissolver.[26]

References

1. *Reactor Fuel Process.*, 8(1): 8 (Winter 1964–1965).
2. V. P. Caracciolo and F. G. Rust, Alniflex Process for Dissolving Zirconium–Uranium Alloy in 304L Stainless-Steel Vessels, *Ind. Eng. Chem., Process Design Develop.*, 5(4): 364-368 (1966).
3. *Power Reactor Technol. Reactor Fuel Process.*, 10(1): 64 (Winter 1966–1967).
4. *Reactor Fuel Process.*, 7(4): 231 (Fall 1964).
5. W. W. Schulz, Aqueous Decladding and Dissolution of Plutonium Recycle Test Reactor Fuels. Part 2, PuO_2–UO_2 Fuels, USAEC Report BNWL-204, Pacific Northwest Laboratory, November 1966.
6. *Reactor Fuel Process.*, 8(1): 10 (Winter 1964–1965).
7. R. G. Butzman and B. J. Newby, Laboratory Feasibility Studies of Aqueous Reprocessing of Zirconium Dioxide–Uranium Dioxide Fuels, USAEC Report IN-1069, Idaho Nuclear Corporation, March 1967.
8. *Reactor Fuel Process.*, 9(2): 69 (Spring 1966).
9. J. A. Buckham, Idaho Nuclear Corporation, personal communication, April 1967.
10. *Reactor Fuel Process.*, 8(1): 12 (Winter 1964–1965).
11. *Reactor Fuel Process.*, 8(3): 147 (Summer 1965).
12. L. A. Decker and R. R. Hammer, A Study of Three Methods for Reprocessing Graphite Fuels: Mixed-Acid Oxidation, Grind-Leach, and Electrolytic Disintegration, USAEC Report IN-1018, Idaho Nuclear Corporation, January 1967.
13. *Reactor Fuel Process.*, 9(3): 151 (Summer 1966).
14. D. G. Karraker and W. C. Perkins, Recovery of Actinides Produced by a Nuclear Detonation in Volcanic Tuff or Desert Alluvium, USAEC Report DP-1055, Savannah River Laboratory, December 1966.
15. *Reactor Fuel Process.*, 7(2): 97 (Spring 1964).
16. W. W. Schulz, Trilaurylamine Extraction of Neptunium and Plutonium from Purex Process Waste, *Ind. Eng. Chem., Process Design Develop.*, 6: 115-121 (1967).
17. *Reactor Fuel Process.*, 8(4): 198 (Fall 1965).
18. D. L. West and R. Narvaez, Curium Process Development. III-2. Identification of Solvent Degradation Products, USAEC Report DP-1016, Savannah River Laboratory, November 1966.
19. *Reactor Fuel Process.*, 9(1): 19 (Winter 1965–1966).

20. *Reactor Fuel Process.*, 9(3): 150 (Summer 1966).
21. *Reactor Fuel Process.*, 9(2): 71 (Spring 1966).
22. D. Brown, S. N. Dixon, and P. J. Jones, The Routine Recovery and Purification of Protactinium-231, *J. Inorg. Nucl. Chem.*, 28: 2529-2533 (1966).
23. J. D. McIntosh and C. J. Raab, Remote Maintenance in a Large Scale Separations Plant, USAEC Report ISO-SA-25, Isochem, Inc., July 15, 1966. Also appeared in *Proceedings of the 14th Conference on Remote Systems Technology*, Oct. 30–Nov. 1, 1966, pp. 196-202, American Nuclear Society.
24. J. A. Buckham, Idaho Nuclear Corporation, Chemical Technology Branch, personal communication, Mar. 1, 1967.
25. *Power Reactor Technol. Reactor Fuel Process.*, 10(2): 152 (Spring 1967).
26. C. A. Lorenzen, Development of a Continuous Dissolver for Plutonium Scrap, USAEC Report ISO-SA-29, Isochem, Inc., Oct. 7, 1966. Also presented at the Pacific Northwest Regional Meeting, American Institute of Chemical Engineers, Richland, Wash., Oct. 27–29, 1966.

Progress in Waste-Disposal Research and Development

By Phillip Fineman

Treatment of Intermediate-Level-Activity Wastes

INCORPORATION IN ASPHALT

Oak Ridge National Laboratory (ORNL) is carrying out development work on the asphalt-immobilization of intermediate-level-activity wastes (ILW).[1] As part of this work, ORNL has issued a recent report[2] that presents a preliminary design and cost estimate of a full-scale plant that will be used for treating concentrated, ORNL-produced ILW. In the ORNL process (Fig. 1), concentrated ILW and emulsified asphalt (type RS-2)* are mixed in a wiped-film evaporator† operating at 320° F, where water is evaporated from the asphalt–ILW mixture. The product, a homogeneous mixture of salts and asphalt, is collected in 55-gal steel drums. The drums after being sealed are suitable for long-term storage or burial. The evaporator condensate is sampled and routed to the ORNL low-level-activity waste system. Noncondensable gases are passed through an absolute filter prior to entering the plant off-gas system.

For a plant designed to process 400,000 gal of ILW per year, the total capital cost for building and equipment (Fig. 2) was estimated to be about $331,000. The operating cost was estimated at $0.34 per gallon, assuming 20-year amortization of capital without interest, and $0.37 per gallon, assuming 20-year amortization with 4% interest.

Conversion of High-Level-Activity Wastes to Solids

Three waste-solidification processes will be tested on an engineering scale (using plant-level-activity wastes) in a demonstration program at Pacific Northwest Laboratory (PNL).[4] The three selected processes are: the PNL spray-solidification process, the ORNL pot-calcination and rising-level glass processes,[5] and the BNL (Brookhaven National Laboratory) phosphate-glass process.[6] These processes will be demonstrated in a multipurpose facility, known as the Waste Solidification Engineering Prototype (WSEP). The main process features[7] of the WSEP are shown in Fig. 3. Recently reported PNL tests[8] of these processes conducted with simulated Purex-process wastes (PW) are discussed later.

RADIANT SPRAY CALCINER AND MELTER

The spray solidification of wastes is carried out by means of a radiant-heat spray calciner coupled to a continuous melter.[9] Toward the end of a spray-calciner run (Run CSCM-8‡),[10] a crack was discovered in the melter fabricated of Pt–0.5 wt.% Rh.[8] The crack, which was star shaped and slightly longer than 1 in., was at the knuckle radius of the 80-mil-thick bottom head of the melter. The failure occurred after about 440 hr of heated service (900 to 1250°C). Highlights are presented of the investigation into the cause of the failure. The failure was determined to be a brittle fracture. Although the exact cause has not been firmly established, present information indicates embrittlement was caused by alloying under locally reducing conditions (such as from inadvertent additions of stainless steel into the melt). It is known, for example, that a phosphorus content[11] of as little as 30 ppm in platinum will reduce its high-strength factor by a factor of 2. The failure may have been accelerated by the relatively high rate of grain growth; by the entrance of melt into enlarged grain boundaries that wedge the grains further apart

*Type RS-2 asphalt[3] is a rapidly settling, high-viscosity, emulsified asphalt. It contains 63% base asphalt, 35% water, and 2% anionic emulsifying agent.

†A wiped-film evaporator is an agitated, thin-film evaporator. These evaporators are capable of handling liquids that are highly viscous, have low thermal conductivity, and contain salts that may crystallize during evaporation or are heat sensitive.

‡In this run, PW-1 waste was modified by the addition of sodium, lithium, and phosphate to a ratio of metallic cation (MC) to phosphorus (P) of 2.75. This produced a low-melting-point (880°C) product. The PW-1 wastes are high in nitric acid (5*M*), free of sulfate ion, and contain added iron from a dissolvable container used to transport chopped fuel.

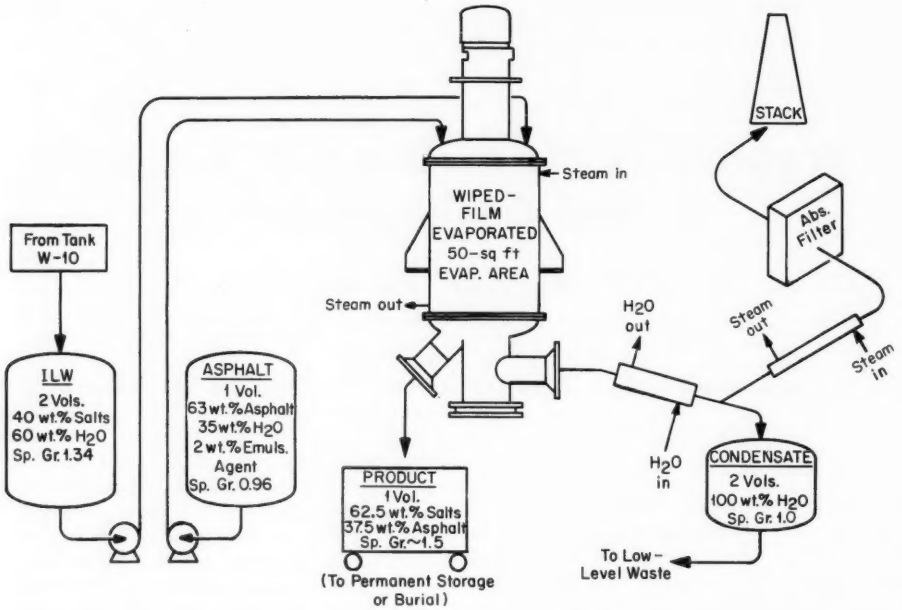


Fig. 1 Schematic flow sheet of ORNL process for asphalt-immobilization of concentrated intermediate-level-activity aqueous wastes (ILW).[2]

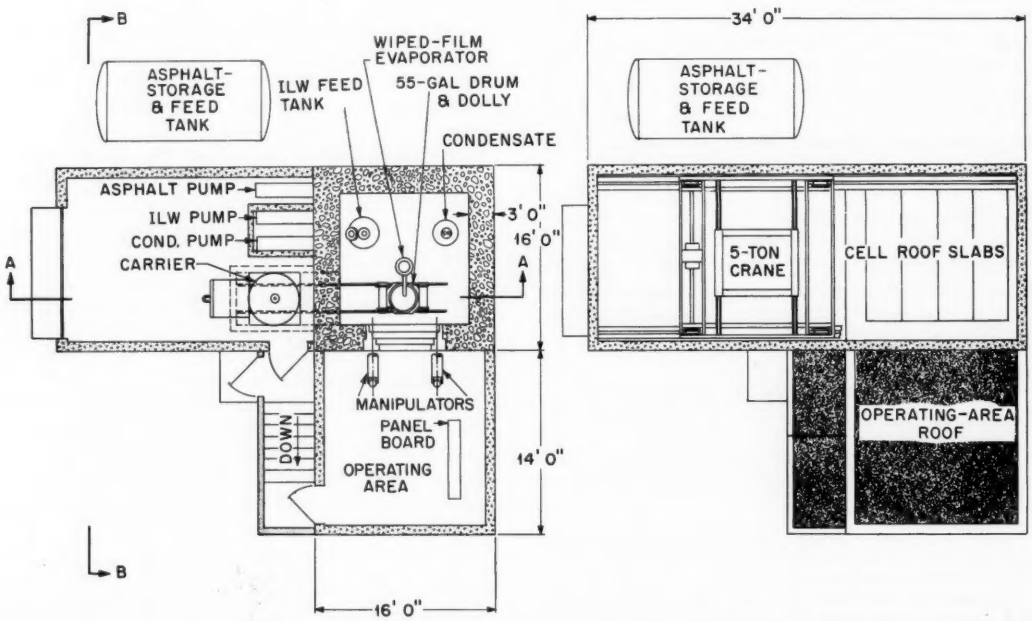


Fig. 2 Plan views of ORNL plant for asphalt-immobilization of ILW.[2]

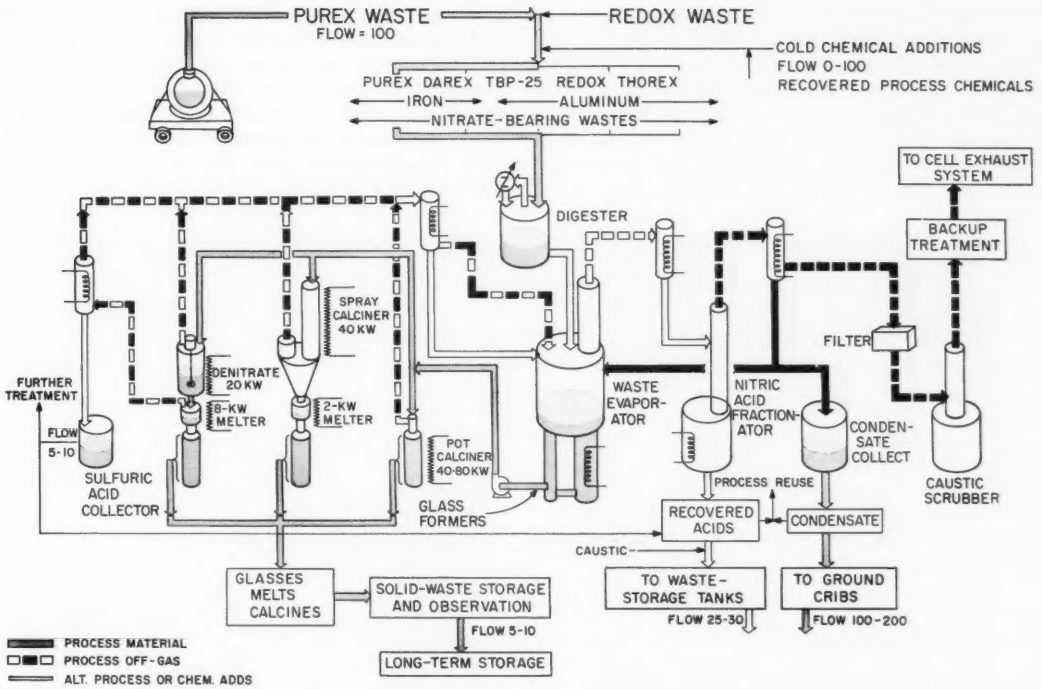


Fig. 3 Waste solidification engineering prototype (WSEP): schematic flow diagram.[7]

due to a combination of thermal, pressure, and stress gradients; or by both.[8] Since test results on the failed material were inconclusive, a laboratory program is under way to simulate the suspected cause. Meanwhile the melter vessel has been replaced with an all-platinum unit of similar design.

POT SOLIDIFICATION PROCESSES—RISING-LEVEL POTGLASS PROCESS

Pot solidification processes produce solidified waste in a cylindrical vessel (pot) suitable for permanent storage. The rising-level (RL) potglass process is one of three pot processes developed at ORNL. In this process the molten-phase level rises throughout the filling operation. Additives necessary to form a final product of glass or ceramic are added to the waste in either the feed line or the pot. In a recently reported run,[8] which was made with PW-1 feed, the compositions of the feed stream and pot were adjusted chemically to a final M-to-P ratio of 2.9. Sodium and phosphate ions were added to the feed stream; lithium and phosphate ions were added to the pot. In this way there was always a meltable mixture in case of poor proportioning of either stream. Also, the use of a separate pot additive stream was desirable to reduce precipitation of solids in the feed tank. The run was terminated after 29 hr when corrosive failure of the bottom of the pot (310 stainless steel, $5/16$-in.-thick wall) took place. Inspection of the pot in the vicinity of the melt showed pot corrosion rates of 100 to 200 mils/day (pot wall temperature of 950°C) and complete loss of pot thermocouple wells (304L stainless steel; $3/4$-in., schedule-80 pipe). Although earlier static laboratory tests[12] indicated that the corrosion rate for the 304L stainless steel would be excessive, these same tests showed that the corrosion rates for the 310 stainless steel should have been much lower than that attained in the reported run. An understanding of this discrepancy in test results is being sought.

The molten waste (30 liters) that leaked from the failed pot was safely contained by the furnace susceptor without damage to any equipment. The furnace was quickly restored to operating service.

PHOSPHATE-GLASS PROCESS

High-level radioactive aqueous wastes are converted to a phosphate glass by adding phosphoric acid to the waste and heating the solution in two steps to 1100 to 1200°C. The glass is produced directly and continuously without an intermediate drying step. A test of this process was successfully completed.[8] In the initial portion of the run, the feed was a PW-2

waste,* and, near the midpoint of the run, the feed was changed to a PW-1 waste. After adjustment of the two feeds with phosphoric acid, the ratio of M to P was 2.09 for the PW-2 waste and 1.00 for the PW-1 waste. Normal operation of the process during the run was good. Improvements are planned in the control of the denitrator–evaporator solution temperature and the feeding from the evaporator to the melter.

The product receiver, a 12-in.-diameter mild-steel vessel, was continuously filled to a 22-in. melt level. No stalagmite formations were observed at filling rates ranging from 12 to 28 liters/day ($\frac{1}{2}$ to 1 cu ft/day). The solidified product in the pot was found to be completely devitrified. However, samples (quick chilled) taken during the run were quite glassy. An overall concentration factor (adjusted feed to product) of 7.6 was attained.

No observable oxidation or corrosion occurred on the inner wall of the product receiver in either the above-melt or in-melt regions. The oxidation corrosion on the outer wall was about 11 mils. This corrosion rate is equivalent to 6.7 mils/day and may be compared to the 1.3 mils/day obtained when a similar receiver in an earlier run was blanketed with a flowing stream of nitrogen.[13]

General

MANAGEMENT OF HIGHLY RADIOACTIVE SOLID WASTES FROM THE EBR-II FUEL CYCLE FACILITY

A recent open-literature article[14] describes management of high-level, noncombustible radioactive solid wastes produced in fuel-recovery and refabrication operations at the Fuel Cycle Facility (FCF)† of the Experimental Breeder Reactor II (EBR-II) complex. The EBR-II complex was built at the National Reactor Testing Station, Idaho. In the FCF, which is adjacent to the EBR-II reactor building, the remote reprocessing and refabricating of the discharged reactor fuel is accomplished. This arrangement permits the reactor to operate on a closed fuel cycle.

The FCF consists of two heavily shielded cells that are connected to each other by transfer locks for material and equipment transfer. One cell contains an air atmosphere (Air Cell) where the discharged fuel subassemblies are dismantled or assembled. The other cell, which adjoins the Air Cell, has an argon atmosphere (Argon Cell). In this cell fuel processing and fabrication are carried out.

Typical solid wastes generated in FCF operations consist of the following: stainless-steel scrap materials from the dismantling of the EBR-II fuel and blanket subassemblies; stainless-steel-cladding scrap from the decanning of the fuel elements; used zirconium oxide crucibles, fume traps, and graphite molds from the fuel reprocessing step; used graphite crucibles from the fuel refabrication step; and discarded equipment items. These wastes must be disposed of on a routine basis.

Four sizes of steel containers (Fig. 4) are used for packaging the wastes; these consist of small pails 10, 12, or 24 in. high by $10\frac{1}{4}$ in. in inside diameter and a large container 6 ft high by 11 in. in inside diameter. The small pails are used in the Argon Cell for packaging of the stainless-steel scrap from the fuel elements and also for packaging of fume traps, molds, and crucibles. The filled pails are capped in the Argon Cell and transferred to the Air Cell where they are inserted into the 6-ft-high container. The 6-ft-high containers are also used for packaging of fuel-subassembly hardware and blanket elements.

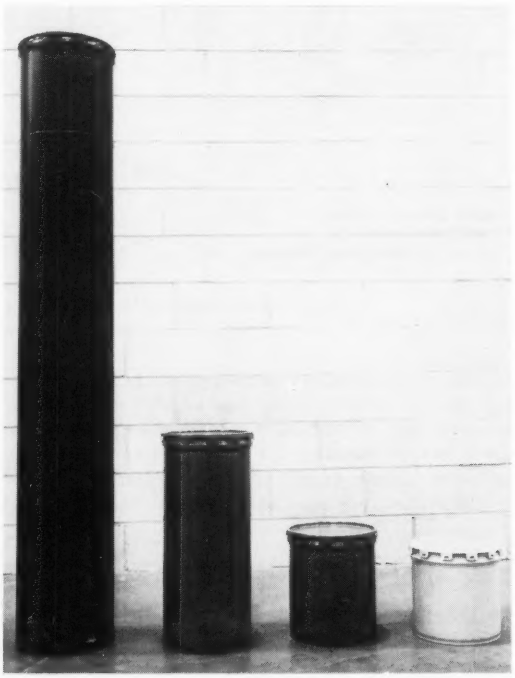


Fig. 4 Four sizes of steel containers used for packaging of FCF solid wastes.[14]

*A PW-2 waste is a combined first- and second-cycle Purex-process waste. It is high in acid (5*M*), contains iron and sulfate ions from the ferrous sulfamate reductant, but does not include iron from the dissolvable container.

† An extensive description of this facility is given in the Fall 1964 issue of *Reactor Fuel Processing*.[15] Operations conducted in the facility are reviewed in other issues of *Reactor Fuel Processing*.[16]

The 6-ft-high containers are capped in the Air Cell and transferred one at a time to a waste-handling coffin (Fig. 5). The coffin is a vertical, shielded ($8\frac{3}{4}$ in. of lead) cylindrical container; it accommodates one waste container that is loaded through the top of the coffin and discharged from it via the bottom.

Operation of the facility is based on a philosophy of long-term interim storage of packaged wastes rather than one of permanent burial of these wastes. It is assumed that all wastes will still be radioactive at the end of 5 years; therefore high-integrity liners are used in the burial holes. (The holes are located on 6-ft centers at a distance of 12 ft between rows.)

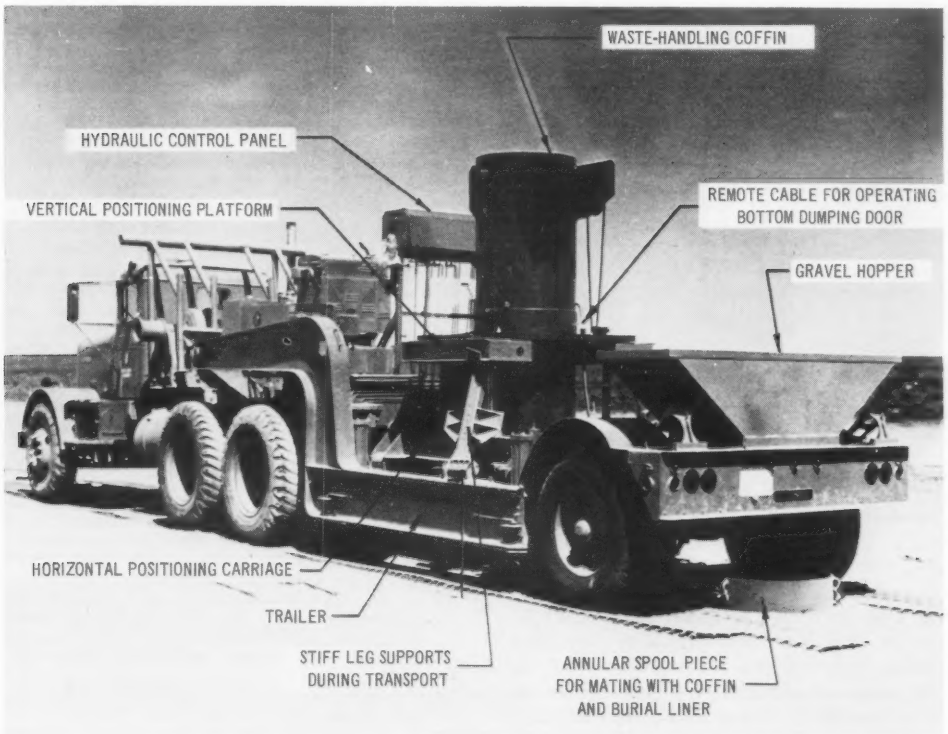

WASTE-HANDLING COFFIN
HYDRAULIC CONTROL PANEL
VERTICAL POSITIONING PLATFORM
REMOTE CABLE FOR OPERATING BOTTOM DUMPING DOOR
GRAVEL HOPPER
HORIZONTAL POSITIONING CARRIAGE
TRAILER
STIFF LEG SUPPORTS DURING TRANSPORT
ANNULAR SPOOL PIECE FOR MATING WITH COFFIN AND BURIAL LINER

Fig. 5 Waste-handling coffin shown on specially constructed lowboy trailer.[14]

A special lowboy trailer was built (Fig. 5) to transport the waste-handling coffin from the FCF to the burial facility. The trailer allows the coffin to be positioned over the burial hole (Fig. 6) and the waste container to be discharged with a minimum amount of radiation hazard to operating personnel. The trailer has a hopper for holding gravel which serves both as backfill and shielding for the burial hole.

The burial facility is a controlled-access, fenced area (410 by 370 ft), approximately one-half mile from the EBR-II complex. The facility is located in no known water-flow patterns; however, the area was built up by banking the earth to a level several feet above the original ground level to provide surface runoff away from the burial facility and to eliminate chances of flooding under any foreseeable conditions.

If in the future it should become desirable to do so, these liners, together with the packaged wastes, could be removed for burial elsewhere. The liners are made of schedule-10 steel pipe (12 ft 4 in. long by 16 in. in outside diameter with a $\frac{1}{4}$-in.-thick wall); they are closed at the bottom end and provided with a top closure plate. The top plate is welded on after the 6-ft waste container has been dropped into the liner and covered with gravel.

The first one hundred 6-ft waste containers that were buried contained an estimated 6600 kg of waste material with an estimated 1.6×10^6 curies of activity (at the time of filling of the containers). The kinds and amounts of radioactivity associated with the waste are dependent upon operation of the reactor. Forty-one of these containers were filled with stain-

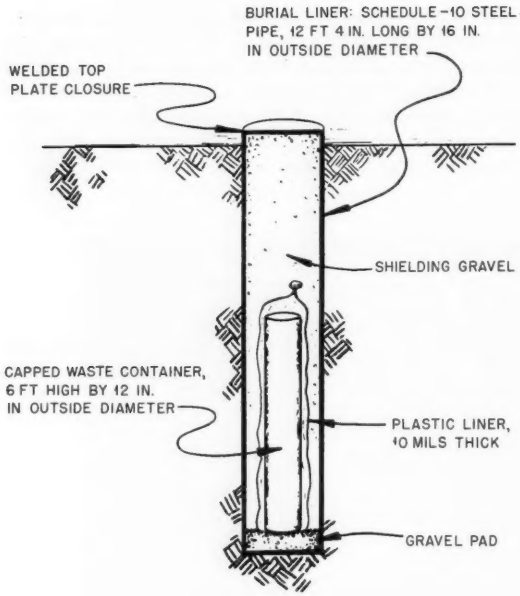

BURIAL LINER: SCHEDULE-10 STEEL PIPE, 12 FT 4 IN. LONG BY 16 IN. IN OUTSIDE DIAMETER
WELDED TOP PLATE CLOSURE
SHIELDING GRAVEL
CAPPED WASTE CONTAINER, 6 FT HIGH BY 12 IN. IN OUTSIDE DIAMETER
PLASTIC LINER, 10 MILS THICK
GRAVEL PAD

Fig. 6 Schematic diagram of burial hole in EBR-II burial facility.[14]

less-steel scrap and discarded axial and radial blanket elements from the dismantling of the subassemblies in the Air Cell. These containers varied in weight from 18 kg to 159 kg and had estimated curie contents of from 1500 to 58,000. Their radiation levels were estimated to be 95 to 200,000 r/hr at 1 ft in air. The other fifty-nine 6-ft waste containers, filled with the processing scrap from the Argon Cell, averaged about 55 kg each and had estimated curie contents of 1000 to 25,000. The radiation levels of these waste cans were estimated to be 50 to 900,000 r/hr at 1 ft in air. These wide ranges of curie amounts and radiation levels are the result of the wide range of burnup levels (0.1 to 1.2 at.%) of the fuel that was sent to the FCF for processing.

At the time of burial, the cooling time for the isotopic wastes will have varied from 20 days to 90 days with isotope half-lives ranging from 8 days to 28 years. Radiation measurements were taken at the burial facility after the container was dropped in the hole and the gravel shielding backfill was placed; they were less than 1 mr/hr at 6 in. from the top of the liner.

References

1. *Power Reactor Technol. Reactor Fuel Process.*, 10(2): 176 (Spring 1967).
2. A. M. Rom, Incorporation of Intermediate-Level Waste in Asphalt: Preliminary Design and Cost Estimate of a Full-Scale Plant for ORNL, USAEC Report ORNL-TM-1697, Oak Ridge National Laboratory, January 1967.
3. Federal Specifications, Asphalt Paving Emulsions, SS-A-674b, Nov. 25, 1952, and Amendment-1, Sept. 2, 1958.
4. *Reactor Fuel Process.*, 9(4): 230 (Fall 1966).
5. *Reactor Fuel Process.*, 9(4): 228 (Fall 1966).
6. *Reactor Fuel Process.*, 9(4): 229 (Fall 1966).
7. W. H. Regan (Ed.), *Proceedings of the Symposium on the Solidification and Long-Term Storage of Highly Radioactive Wastes*, February 14–18, 1966, Richland, Washington, USAEC Report CONF-660208, p. 454, November 1966.
8. A. M. Platt (Ed.), Fixation of Radioactive Residues, Quarterly Progress Report, Research and Development Activities, July–September 1966, USAEC Report BNWL-365, pp. 11-33, Pacific Northwest Laboratory, March 1967.
9. *Power Reactor Technol. Reactor Fuel Process.*, 10(1): 89-90 (Winter 1966–1967).
10. A. M. Platt (Ed.), Fixation of Radioactive Residues, Quarterly Progress Report, Research and Development Activities, April–June 1966, USAEC Report BNWL-306, p. 11, Pacific Northwest Laboratory, August 1966.
11. A. Jedele, *Z. Metallk.*, 27: 27 (1935).
12. A. M. Platt (Ed.), Fixation of Radioactive Residues, Quarterly Progress Report, Research and Development Activities, July–September 1966, USAEC Report BNWL-365, pp. 4-6, Pacific Northwest Laboratory, March 1967.
13. *Power Reactor Technol. Reactor Fuel Process.*, 10(2): 177 (Spring 1967).
14. W. F. Holcomb, D. M. Paige, C. E. Stevenson, and E. D. Graham, High-Level Radioactive Solid Waste Burial Operations for the EBR-II Fuel Cycle Facility, *Nucl. Eng. Design*, 5: 221-227 (1967).
15. *Reactor Fuel Process.*, 7(4): 250-254 (Fall 1964).
16. *Reactor Fuel Process.*, 8(1): 27-28 (Winter 1964–1965); 8(2): 97-99 (Spring 1965); 8(3): 162-163 (Summer 1965); 8(4): 214-215 (Fall 1965); 9(1): 32-33 (Winter 1965–1966).

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